

# Galvanic Corrosion Behaviour of Aluminium 3004 and Copper in Tropical Marine Atmosphere

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**Abstract:** The galvanic corrosion behaviour of aluminium 3004 and copper with different area ratios were studied in the tropical marine atmosphere at Tuticorin harbour over a period of 426 days. The area ratios of  $A_{Al} : A_{Cu}$ , studied were 1:1, 1:2, 1:4, 1:8, 2:1, 4:1 & 8:1. The galvanic corrosion behaviour of metals was studied in terms of relative increase in the corrosion rate of aluminium due to galvanic coupling with copper, relative decrease in the corrosion rate of copper due to galvanic coupling with aluminium, and the susceptibility of aluminium to pitting owing to galvanic coupling with copper. The galvanic potential and galvanic current of the system were monitored. Pits of different dimensions ranging from mild etchings to perforations were experienced on the borders and the surfaces of the interface of aluminium in contact with copper. The weathering parameters and the environmental pollutants which have a major role in influencing the galvanic corrosion of metals were also monitored. The corrosion products resulting from galvanic corrosion were analysed using XRD and the pitting on aluminium resulting from galvanic corrosion has been highlighted in terms of pit depth, size and density of pit, using a high resolution microscope.

**Keywords:** aluminium 3004; copper; galvanic corrosion; corrosion products; pitting; tropical marine atmosphere

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## 1 Introduction

When two dissimilar conducting materials in electrical contact with each other are exposed to an electrolyte, a current, called the galvanic current, flows from one to the other. Galvanic corrosion is that part of the corrosion that occurs at the anodic member of such a couple and is directly related to the galvanic current by Faraday's law. Galvanic corrosion occurs commonly in atmospheric environments as different combinations of materials are used in buildings and structures exposed to indoor and outdoor atmospheres. A test program of galvanic corrosion in atmospheres was started as early as 1931 by the American Society for Testing and Materials (Kucera and Mattsson, 1982). Since then, a number of extensive exposure programs have been carried out all over the world (Rosenfeld, 1962; Doyle and Wright, 1988; Compton and Mendizza, 1955; Dey *et al.*, 1966). The

various aspects of atmospheric galvanic corrosion have been discussed in a comprehensive review by Kucera and Mattsson (1982).

Military materials are deployed and used worldwide under varied and severe environmental conditions. In the design and fabrication of equipment the use of electrochemically similar metal is desirable, but is not feasible in many instances when structural, electrical and other important design criteria are taken into account. Consequently under such conditions, especially in marine atmospheres, significant galvanic corrosion problems are encountered. Although the danger of galvanic corrosion has been recognized generally, very little work has been done to quantitatively measure the extent of corrosion of various galvanic couples, and use of such data as the basis for ranking of galvanic couples. The magnitude of galvanic corrosion depends not only on the potential difference of dissimilar metals, but also on the kinetic parameters such as corrosion rates and area ratios (Mansfeld, 1971; LaQue, 1951). Though measurements of electrode potential indicate the protective nature of the corrosion products (Vassie and McKenzie, 1985), firsthand knowledge on galvanic current and weight-loss data, throw more light on the galvanic interactions of the metals in contact (Mansfeld and Kenkel, 1975a). Pitting is the form of localized corrosion attack usually encountered, when dissimilar metals are in galvanic contact. By virtue of environmental pollutants and weathering conditions (Campbell, 1950), this can lead to premature failure of the systems in operation.

There has been a need for the specific data relating to galvanic interactions of certain metal combinations frequently encountered in military equipment in tropical marine atmosphere to aid the design engineer in the selection of metal combinations to achieve maximum corrosion resistance. Galvanic corrosion studies of various metals & alloys under lab conditions using NaCl solution have been well documented (Mansfeld, 1971; Mansfeld and Kenkel, 1975a, 1975b & 1975c; Mansfeld *et al.*, 1974; Walker, 1979). Ha *et al.* (2004) studied the galvanic corrosion behaviour of aluminium & copper sleeves in power distribution systems, in NaCl solution by electrochemical techniques. Aluminium is used excessively in the modern world, and the uses of the metal are extremely

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diverse due to its many unusual combinations of properties. Applications for aluminium include ship hulls, piers, tank interiors, offshore structure, submerged pipelines and piling (Musa *et al.*, 2011a). Copper is a very widely used material for its excellent electrical and thermal conductivities in many industrial applications (Mihit *et al.*, 2006; Musa *et al.*, 2011b; Musa *et al.*, 2011c). Understanding the corrosion process will help in reducing the safety and economic impact of corrosion damage. Aluminium and copper are both widely used conductor materials and each has its own benefits. The metal selected for each application needs to have the necessary mechanical, electrical and chemical properties to fulfil its intended function, while meeting the economic requirements of the installation (Wilhelm, 1988).

Hence in the present study, the galvanic interactions of aluminium (Al) and copper (Cu) have been studied in depth to delineate weight-loss corrosion rate, with respect to varying area ratios, galvanic potential, galvanic current, pitting and corrosion products formed at the interface of bimetallic contact, in the tropical marine atmosphere at Tuticorin harbour, Tamilnadu, India.

## 2 Materials and methods

Commercially available metal sheets such as aluminium 3004 (Mn: 1.05%, Mg: 1.12%, Al: balance) of 1.6 mm thickness and copper (Cu: 99.9% pure) of 3 mm thickness supplied by M/s. Lawrence Metal Industries, Chennai were used in this study.

### 2.1 Coupon preparation

The sheets of aluminium and copper were cut into the required number of coupons of the following sizes, 38 mm × 25 mm, 50 mm × 38 mm, 75 mm × 50 mm and 150 mm × 100 mm. All the coupons were provided with a central hole of size 10 mm diameter for effecting galvanic contact. The coupons were polished (Champion, 1952), degreased and weighed to an accuracy of  $10^{-4}$  g. Separate set of coupons was prepared for measuring galvanic current of Al 3004-copper couples as described elsewhere (Mansfeld and Kenkel, 1976).

### 2.2 Test site

The offshore platform of CECRI's centre at Tuticorin harbour is located (longitude 78.13° East and latitude 8.8° North) on the southeast coast of India in the Gulf of Mannar. This platform stands on piles in the open sea at a water depth of 6-7 m, which is about 1.5 km away from the shoreline. The standard atmospheric corrosion testing racks erected at the offshore platform were inclined 45° to horizontal. Though the test site is influenced by both southwest and northeast monsoon, relatively higher amount of salt content in the atmosphere is recorded during southwest monsoon. The site is characterized by the prevalence of 63-77% of relative humidity throughout the year.

## 2.3 Experimental techniques

The area ratios of the couple Al 3004-Copper, viz.  $A_{Al} : A_{Cu}$ , studied were 1:1, 1:2, 1:4, 1:8, 2:1, 4:1 and 8:1. Galvanic coupling was effected by placing one over the other (aluminium over copper) using insulated G.I bolt & nut to avoid tri-metallic contact. The contacts in all couples were made possible with the help of G.I bolt & nut to an extent of equal magnitude. Required number of coupons of freely corroding and galvanic couples in triplicate were exposed by mounting on the atmospheric corrosion test racks. In all the couples the aluminium coupon was positioned on the top of the copper coupon to avoid contamination of aluminium by the corrosion products of copper. The galvanic potential values of the systems were monitored with the technique as described elsewhere (Vassie and Mckensie, 1985), using high impedance digital multi-meter with respect to saturated calomel electrode (SCE). The galvanic current was monitored using the atmospheric corrosion monitor as described elsewhere (Mansfeld and Kenkel, 1976) using zero resistance ammeter. The test coupons were removed at the intervals of 133, 214, 302 and 426 days. The gravimetric corrosion rates were calculated after removal of corrosion products using the recommended pickling solutions (ASTM standards, 2003). Weight-loss method was used to calculate the corrosion rate ( $r_{wl}$ ) of metals under galvanically coupled condition, the relative increase in the corrosion rates  $(r_{wl}-r_0)/r_0$  of aluminium due to galvanic coupling with copper and the relative decrease in the corrosion rates  $(r_0-r_{wl})/r_0$  of copper due to galvanic coupling with aluminium, and  $r_0$  the corrosion rate of freely corroding individual metals. The corrosion products were analyzed using X-ray diffractometer (Model-PW3040/60 X'pert PRO). The pitting corrosion behaviour of the galvanically coupled aluminium coupons exposed for 133, 214, 302 and 426 days were recorded in terms of depth of pit, density and size of pits (ASTM standards, 2005), using a high resolution microscope. The pitting factors of the galvanically coupled aluminium coupons of 426 days of exposure were also calculated. The salt content in the air and the meteorological data such as % relative humidity, maximum & minimum temperature, total sunshine hour and rainfall, were recorded periodically over the study period. The salt content in the air was determined by wet candle method, the % relative humidity was monitored using wet & dry bulb thermometers, the maximum & minimum temperature was monitored using maximum minimum temperature system, the rainfall was measured using rain gauge and the sunshine hour was recorded using a calibrated Campbell-Stokes pattern sunshine recorder.

## 3 Results

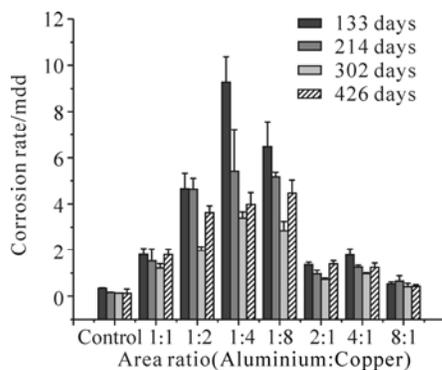
Table 1 shows the meteorological data of the test site. The percent relative humidity of the test site varied from 63.5% to 77.4% and the salt content in the air was found to vary

from 0.42 mdd to 6.48 mdd. Maximum rainfall (172.3-182.5 mm) was recorded during northeast monsoon period (October–December) and intermittent rainfall (2.5-12.9 mm) was recorded from May to September. Maximum sunshine hours (208.8-298.4 h) was recorded between January and March, when there was no rainfall. Fig.1 and Fig.2 portray the corrosion behaviour of aluminium coupled to copper and

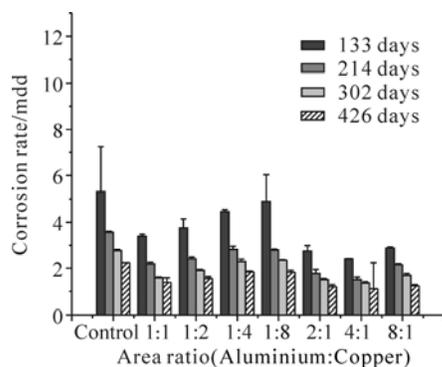
copper coupled to aluminium, respectively. In general, the corrosion rates of aluminium coupled to copper over a wide range of area ratios are higher than that of the freely corroding aluminium, while the corrosion rates of copper coupled to aluminium over a wide range of area ratios are lower than that of the freely corroding copper.

**Table 1 Meteorological data and salt content in the air at the test site**

Period	Relative humidity/%	Temperature maximum/°K	Temperature minimum/°K	Total sunshine/h	Salt content in the air/mdd	Rainfall/mm
Jun'11	72.23	301.33	297.26	190.24	6.06	6.2
July'11	71.24	304.67	299.45	172.20	4.22	8.1
Aug'11	68.72	303.89	298.31	178.00	1.02	6.4
Sep'11	76.25	305.78	301.75	251.00	3.21	12.9
Oct'11	76.75	306.82	298.69	205.20	1.98	172.3
Nov'11	75.38	305.29	297.88	200.70	1.32	182.5
Dec'11	77.26	304.87	298.27	209.50	2.97	176.4
Jan'12	69.26	306.21	297.86	208.80	0.88	3.5
Feb'12	65.26	307.24	297.76	270.30	0.42	Nil
Mar'12	63.45	307.88	297.95	298.40	0.56	Nil
Apr'12	68.28	307.26	300.74	271.30	1.59	Nil
May'12	72.97	305.12	300.82	224.90	0.98	2.5
June'12	76.87	304.78	300.76	200.70	6.48	3.2
July'12	77.35	306.78	300.64	214.70	5.92	Nil



**Fig. 1 Corrosion behaviour of aluminium coupled to copper**



**Fig.2 Corrosion behaviour of copper coupled to aluminium**

Table 2 summarizes the data on relative increase/decrease in the corrosion rate of dissimilar metals of Al-Cu. The relative increase in the corrosion rate of aluminium resulting from galvanic coupling with copper is in the descending order of 1:4 > 1:8 > 1:2 > 1:1 > 4:1 > 2:1 > 8:1, for 133 days of exposure, whereas for 426 days exposure it is in the descending order of 1:8 > 1:4 > 1:2 > 1:1 > 2:1 > 4:1 > 8:1. The relative decrease in the corrosion rate of copper in galvanic contact with aluminium is in the descending order of 4:1 > 2:1 > 8:1 > 1:1 > 1:2 > 1:4 > 1:8, for 133 days of exposure, whereas for 426 days exposure it is in the descending order of 4:1 > 2:1 > 8:1 > 1:1 > 1:2 > 1:8 > 1:4. The galvanic potential and galvanic current density values of Al-Cu couples over the period of exposures are shown in Fig.3 and Fig.4, respectively. The legends C-Al and C-Cu in Fig.3 denote the open circuit potentials (OCP) of freely corroding Al and copper, respectively. The galvanic potential values decreased sharply up to 50 days of exposure and thereafter exhibited nobler direction shift till 190 days. However after 190 days it tended to decrease. During the initial period of exposure (up to 50 days) a slight increase in the galvanic current density was recorded only for the area ratios 4:1 and 1:8. Further for the area ratio 1:8, there was a sharp increase in the galvanic current density after 200 days. The galvanic current density for the area ratio 8:1 showed increasing trend from 190 day onwards.

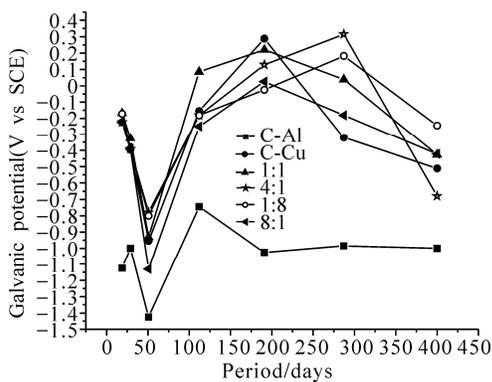
**Table 2** Relative increase/decrease in the corrosion rate of aluminum 3004 and copper of aluminum-copper couple

Area ratio $A_{Al}:A_{Cu}$	133 days		214 days		302 days		426 days	
	Al*	Cu**	Al*	Cu**	Al*	Cu**	Al*	Cu**
1:1	4.2	0.36	8.9	0.39	8.2	0.43	13.9	0.38
1:2	12.3	0.29	28.9	0.33	14.1	0.31	28.8	0.30
1:4	25.4	0.16	33.9	0.21	24.8	0.18	31.7	0.17
1:8	17.4	0.08	32.4	0.22	20.6	0.15	35.7	0.18
2:1	2.9	0.48	5.3	0.50	4.5	0.46	10.4	0.46
4:1	4.1	0.55	7.1	0.58	6.5	0.51	9.2	0.50
8:1	0.5	0.46	3.2	0.40	2.2	0.39	2.5	0.44

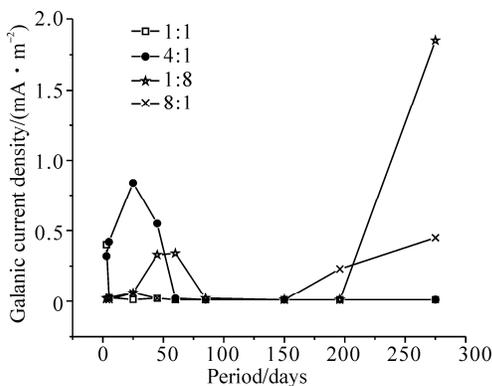
Key to the table:

\* $(r_{wi}-r_0)/r_0$ : relative increase in the corrosion rate of aluminium 3004

\*\* $(r_0-r_{wi})/r_0$ : relative decrease in the corrosion rate of copper



**Fig. 3** Galvanic potential values of aluminium-copper couples



**Fig. 4** Galvanic current density values of aluminium-copper couples

Table 3 summarizes the pitting on aluminium 3004 as a result of galvanic contact with copper. Pitting on aluminium coupled to copper is observed beneath the white corrosion products on the borders and within the interface of the bimetallic contact. The depth of pit varies from mild

etchings to perforations on the surfaces of galvanically coupled aluminium, while the depth of pit ranges between 150  $\mu\text{m}$  and 400  $\mu\text{m}$  on the surfaces of freely corroding aluminium.

Significant variations in the density and size of pit between the surfaces of galvanically corroding aluminium (Maximum no.  $1.0 \times 10^4/\text{m}^2$ , 0.1-24.5  $\text{mm}^2$ ) and freely corroding aluminium (maximum no.  $2.5 \times 10^3/\text{m}^2$ , 0.6-4.0  $\text{mm}^2$ ), are observed for 133 days of exposure. The pit depth of galvanically coupled aluminium ranges between 200  $\mu\text{m}$  and perforation, for 133 days. On the other hand for 426 days exposure, the depth of pit on galvanically corroding aluminium is from 400  $\mu\text{m}$  to perforation, while it is limited to a maximum of 400  $\mu\text{m}$  for the freely corroding aluminium.

Significant variations in the density and size of pit between the surfaces of galvanically corroding aluminium ( $2.5 \times 10^3/\text{m}^2$ - $1.0 \times 10^5/\text{m}^2$ , 0.25-24.5  $\text{mm}^2$ ) and freely corroding aluminium (max. no.  $2.5 \times 10^3/\text{m}^2$ , 0.5 – 2.0  $\text{mm}^2$ ) are observed for 426 days of exposure. In general, perforations are experienced by the aluminium coupled to copper of 1:2 and 1:4 area ratios for 214 days exposure. In 426 days exposure, all the area ratios of aluminium experienced severe pitting to the extent of perforation. Other than tarnishing, no characteristic pitting is observed on the surface of copper coupled to aluminium.

The corrosion product analysis by XRD revealed the presence of the following compounds such as aluminium chloride hexa hydrate,  $\Psi$ -aluminium oxide, aluminium oxide hydroxide,  $\infty$ -aluminium hydroxide, copper (II) chloride di-hydrate & copper (I) chloride, at the interface of the bimetallic contact.

**Table 3 Pitting on aluminium 3004 coupled with copper**

Area ratio $A_{Al}:A_{Cu}$	133 days			214 days			302 days			426 days		
	A	B	C	A	B	C	A	B	C	A	B	C
1:1	1	$1 \geq x = 5$	1	1	$2 \leq x = 5$	2	1	$1 = x \geq 3$	1	2	$1 \leq x = 5$	1-3*
1:2	2	$1 \geq x = 5$	1	2	$2 \geq x = 5$	1-3*	2	$1 \geq x < 5$	1	3	$1 \geq x = 5$	1-3*
1:4	2	$1 \geq x = 5$	1	2	$2 \geq x = 5$	1-3*	3	$2 = x = 5$	1	3	$1 \leq x = 5$	1-3*
1:8	2	$1 \geq x = 5$	1	2	$2 \geq x < 5$	2	3	$1 \geq x < 3$	1	4	$1 \geq x = 5$	1-3*
2:1	1	$1 \geq x = 5$	1	1	$2 \geq x = 5$	2	1	$1 \geq x = 5$	1	2	$1 \leq x = 5$	1-3*
4:1	1	$1 \geq x < 4$	1	1	$2 \geq x = 5$	2	1	$2 \geq x \leq 5$	1	3	$1 \leq x = 5$	1-3*
8:1	1	$2 \geq x < 5$	1	1	$1 \geq x < 5$	1	1	$1 \geq x > 4$	1	1	$1 \leq x = 5$	1-3*
Ctrl	1	$1 = x < 3$	1	1	$1 \geq x < 2$	1	1	$1 < x < 4$	1	1	1-2	1

Key to the table: \*: Perforation; Ctrl: Control

**Table 4 Explanation of some nomenclatures in table 3**

Sl. No.	Density (A)/m <sup>2</sup>	Size (B)/ mm <sup>2</sup>	Depth (C)/mm
1	$2.5 \times 10^3$	0.5	0.4
2	$1.0 \times 10^4$	2.0	0.8
3	$5.0 \times 10^4$	8.0	1.6
4	$1.0 \times 10^5$	12.5	3.2
5	$5.0 \times 10^5$	24.5	6.4

## 4 Discussion

Galvanic corrosion, resulting from a metal contacting another conducting material in a corrosive medium, is one of the most common types of corrosion. Many factors play a role in galvanic corrosion in addition to the potential difference between the two coupled metals. Compared to normal corrosion, galvanic corrosion is generally more complex because, in addition to material and environmental factors, it involves geometrical factors.

The marked decrease in the corrosion rate of aluminium coupled to copper in all the area ratios, excepting the 8:1, over the period of exposure, particularly up to 302 days, is mainly attributed to the formation of corrosion products such as aluminium chloride hexa hydrate,  $\Psi$ -aluminium oxide, aluminium oxide hydroxide,  $\infty$ -aluminium hydroxide, copper (II) chloride di-hydrate and copper (I) chloride at the interface of the bimetallic contact. These products considerably decreased the conductivity of the electrolyte at the interface, leading to less galvanic attack on aluminium (Shreir *et al.*, 1977). The deviation from the exponential decrease in the corrosion rate of aluminium particularly for the 426 days exposure for all the area ratios is mainly due to the higher degree of pitting on aluminium (Table 3). In general, galvanic corrosion in atmosphere is usually restricted to a narrow region of the anode metal near the bimetallic junction because of the high resistance of thin-layer electrolytes formed by rain and water condensation (Rosenfeld, 1962; Zhang, 1998; Zhang and Valeriotte, 1995). The higher corrosion rate experienced by the copper of 1:8 area ratio compared to other area ratios is

mainly due to the fact that the effective area of galvanic contact with aluminium is comparatively less than those of the other area ratios, as is evidenced by the formation of corrosion products such as copper (II) chloride di-hydrate.

The galvanic potential values of the couples reached to a steady state by 125 days of exposure and maintained till 300 days of exposure. Similar observation was earlier made by Foster *et al.* (1996), who reported that the steady-state corrosion potential of a metal electrode depends on whether the surface is active or passive, and the time required for reaching a steady-state value varies with the conditions. The rate of galvanic corrosion may change with time as a result of changes in polarity and in potential difference between the metals in the couple. According to Kucera and Mattsson (1982) galvanic action is most significant in marine atmosphere because of the high conductivity of seawater. Compared to other types of moisture formed under atmospheric conditions, rain is particularly effective in causing galvanic corrosion. In the present study the galvanic corrosion is more severe during the initial exposure period of 133 days. This could be explained that the exposure was commenced during June. The study location usually experiences severe monsoonal wind (southwest) between June and September, which results in higher salt content in the atmosphere, and the intermittent showers of rain during this period resulted in the higher corrosion of metals.

The shifting of galvanic potentials during initial period towards nobler direction is indicative of the protective nature of the corrosion products at the interface of the bimetallic contact, which is important in determining the galvanic corrosion rate (Kucera and Mattsson, 1982). The

extent of pitting experienced by the aluminium coupled to copper of 1:2 and 1:4 area ratios for 214 days exposure and for all the area ratios of 426 days exposure is corroborating with the aggressiveness of the tropical marine atmosphere, prevailed during that period. The pitting factors for 426 days exposure of galvanically corroding aluminium of area ratios, viz. 1:1, 1:2, 1:4, 1:8, 2:1, 4:1 and 8:1 are 2.6, 2.2, 3.1, 2.4, 1.9, 1.8 and 1.8, respectively and for the freely corroding aluminium is 0.8. Thus the durability of aluminium is brought down by a factor of 2 to 4 due to galvanic coupling with copper.

## 5 Conclusions

(1) The relative increase in the corrosion rate of aluminium in galvanic contact with copper can be ranked in the descending order of 1:8 > 1:4 > 1:2 > 1:1 > 2:1 > 4:1 > 8:1, and the relative decrease in the corrosion rate of copper in galvanic contact with aluminium can be ranked in the descending order of 4:1 > 2:1 > 8:1 > 1:1 > 1:2 > 1:8 > 1:4, for 426 days of exposure.

(2) The most favourable area ratios of aluminium-copper in marine atmosphere in terms of weight loss corrosion rate and pitting corrosion are 4:1 and 8:1.

(3) The most unfavourable area ratios of aluminium-copper in marine atmosphere in terms of weight loss corrosion rate and pitting corrosion are 1:1, 1:2 and 1:4.

(4) Pitting corrosion on aluminium in galvanic contact with copper is predominantly decided by the formation of compounds such as,  $\text{Al}_2\text{O}_3$ ,  $\text{AlO}(\text{OH})$  and  $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$  at the interface of aluminium-copper, as influenced by the aggressiveness of the environmental pollutant, caused by the prevailing weathering conditions and monsoon winds.

(5) Aluminium in galvanic contact with copper experienced severe pitting to the extent of perforation, which is predominant as the duration of the exposure extends. In general, it is detrimental to aluminium, when it is in galvanic contact with copper in marine atmosphere.

(6) For 426 days exposure, the pitting factors of aluminium in aluminium-copper, for the area ratios, viz. 1:1, 1:2, 1:4, 1:8, 2:1, 4:1 and 8:1 are 2.6, 2.2, 3.1, 2.4, 1.9, 1.8 and 1.8, respectively. Thus the durability of aluminium is brought down by a factor of 2 to 4 due to galvanic coupling with copper.

(7) The shifting of galvanic potentials towards nobler direction is indicative of the protective nature of the corrosion products at the interface of the bimetallic contact.

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## Author biographies



**G. Subramanian** is currently holding the post of senior principal scientist and head of OPMEC, CECRI Unit, Tuticorin. He has a Ph.D in Marine Sciences and his research since 1983 has focused on a range of topics including atmospheric corrosion, evaluation of paints and coatings, marine biofouling prevention and corrosion in seawater. Besides well cited publications, he has seven Indian patents and one US patent to his credit. He has also commercialized a process.



**S. Palraj** is currently holding the post of principal technical officer at CSIR-CECRI, Karaikudi. He has a Ph.D in Chemistry and has been working in the field of Corrosion Testing & Evaluation for the last 31 years. Presently he is involved in the development of antifouling coatings. Besides well cited publications, he has eight Indian patents and one US patent to his credit. Further he has developed 4 processes and commercialized them.



**S. Palanichamy**, Senior Scientist, holds a Ph.D degree in marine sciences at CSIR-CECRI, Tuticorin. He was instrumental in establishing a strong database on chemical oceanographic features of coastal ocean waters, including the Tuticorin and Mandapam regions. Simultaneously he also investigated the effects of water chemistry and local pollution on corrosion and biofouling phenomena. Currently he is developing antifouling formulations from marine natural products.