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Original article

Role of environmental particulate matters on corrosion of copper

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ABSTRACT

Effect of environmental particulate materials on kinetics and mechanism of corrosion of industrial grade copper exposed in different parts of India is investigated. It is observed that the level of particulate materials in the atmosphere has more dominant role than the acidic gases on initiation of corrosion, formation of protective patina on the surface of the exposed samples leading to mitigation of corrosion. The identification of corrosion products formed on the surface of exposed samples by Raman spectroscopy provides very vital information to explain the observed corrosion rate of the metal computed in different environments. Electrochemical anodic polarization of the exposed samples supports the mechanism proposed for accelerating and protective effect on corrosion of the metals exposed in different environments.

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1. Introduction

Copper and its alloys are very important functional materials widely used in different industrial and household applications. Many existing old heritages available around the world are also made from this metal and are still in intact condition without any major maintenance. The Statue of Liberty, in New York City which has 80 metric tons of copper sheeting is an example of superb protective property of the patina formed on the surface of copper metal. High electrical conductivity of the metal makes it a choice material for transmission of electricity. Due to its low corrosion rate, aesthetic look and also antimicrobial properties, copper roofing has become very popular in buildings of public facilities, residential complexes etc. The metal although very resistant to corrosive attack of normal unpolluted environments, it is quite vulnerable to different types of pollutants present in atmosphere leading to various types of corrosion such as uniform, tarnishing, pitting etc. Owing to its industrial, historical, cultural and economical importance, very extensive studies had been performed in the past to understand the mechanism of corrosion of

copper and its alloys (Leidheiser, 1974; Rozenfeld, 1972; Uhlig, 1948; Aziz and Godard, 1959; Shreir, 1976). It is suggested that stability of corrosion products formed on surface of exposed metal determines its long term rate of corrosion. In neutral and non-polluted environments, a thin layer of Cu_2O and CuO develops on the surface of copper metal. Various pollutants in the environment such as SO_2 , NO_x , salinity and suspended particulate matters (SPM) etc destabilize the protective oxide film formed on the metal surface. Literature survey reveals that considerable efforts had been in the past to correlate the corrosion rate of the metal with atmospheric pollutants present in the atmosphere. Majority of these studies describe the role of sulphur dioxide, hydrogen sulphide and NO_x gases on the corrosion of the metal (Vera et al., 2007, 2008; Kitase et al., 2001; Strandberg, 1998; Salas et al., 2016).

Suspended particulate matters are formed in the environment due to burning of fossil fuels, woods, agricultural produces, dusts etc and deposit on surfaces of metals and materials. Many heavy metals in the form of their oxides, sulphates, nitrates etc are present in these particulate materials. These materials varying in their sizes (micro metre to nanometres) quickly adsorb/physically deposit on surfaces of metals and alloys. Due to hygroscopic nature of salts present in the particulate materials they act as good reservoir of humidity and keep the metals surfaces wet for longer duration of time than the cleaned surface. This facilitates the formation of electrochemical cells and accelerates the corrosion reactions. A

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clean surface of copper metal is very prone to corrosion once wet particulate materials laden with heavy metal salts deposit on them. In comparison to the other metals and alloys, copper forms variety of complexes with different types of anions and its corrosion rate is strongly influenced by the solubility of these complexes in rain water. Literature search reveals that although the information on effect of deposited particulate materials on corrosion of stainless steel, zinc, Al etc are available (Button and Simm, 1985; Grau-Bove and Strlic, 2013; Vernon, 1949; Greenblatt, and Pearlman, 1962; Sanyal and Singhania, 1956) but their role on copper metal is very scarcely available. The present investigation therefore was taken up to study the kinetics and understand the mechanism of corrosion of this metal in different environments where normal constituents responsible for corrosion, namely SO₂ and NO_x were comparable but greatly differed in their content of particulate matters.

2. Experimental procedure

Industrial grade copper metal (97% Cu + 3% Zn), in sheet form was used to generate the data. The metal coupons of size 150 mm × 100 mm × 1 mm were hexa – cut from sheets. The surface of the test panels were first abraded on polishing belt followed by hand polishing using coarse and fine grades of emery papers (100–1200). After degreasing with acetone the samples in triplicate were exposed in atmosphere of various cities of India namely Jamshedpur, New Delhi and Kolkata for duration of 1 and 2 years. The procedure of exposure and determination of the corrosion rate was followed as described in ASTM G-50-10 (2015). The entire test samples were exposed on steel racks fixed at an angle of 45° facing south. To avoid galvanic contacts the samples were fixed to the rack with the help of porcelain insulators and brass hexagonal nut and bolts. The racks were installed at the roof top of buildings at heights of about 10 metres. Care was taken to avoid any shed due to trees and buildings closer to the exposure locations. After the exposures of 1 and 2 years, three sets of the samples were taken out from the racks and brought in the lab to perform different type of tests. The corrosion products and passive film formed on the surface of the specimens were characterized by Raman spectroscopy. To determine corrosion rate the corrosion products formed on the surface of the specimens were cleaned as described in ASTM G1-90 (re-approved 2010). Average corrosion rate of three samples were determined. The uniformity in the corrosion rates of the three identical samples were appreciably good and maximum variation was 0.72% from samples to samples. The data were generated by performing three types of tests. The first sets of experiments were the exposure of samples at different locations of India as stated above. These sites were chosen in view of the fact that they represented atmospheres where gaseous level of pollution in the air was comparable but greatly differed in their PM₁₀ contents. The average annual data for SO₂, NO₂ and PM₁₀ during the exposure period were generated by the Central pollution Control Board of India and are recorded in Table 1. Since corrosion rate of the exposed panels was determined after one and two years of exposure at different sites, it was more pertinent to present pollution data as average yearly than the monthly.

The second set of experiments was performed to characterize the corrosion products by Raman spectroscopy. Almega micro Raman Spectroscope was used by exciting the laser beam of Nd-YAG of 532 nm wavelength on the samples. The power of the laser was kept lowest possible (6 mw) to avoid the transformation of corrosion products due to heating effect of laser. The locations of the specimens to be studied were focused through an Olympus microscope at the magnification of 50. The sample holder had motorized platform with Jokey to have a fine focussing at a suitable desired part of the sample. The grating was 672 lines/mm, 25 μm

Table 1

SO₂, NO₂ and PM₁₀ data generated during the period of exposure tests by Central Pollution Control Board, Delhi at exposure sites of India.

Exposure sites	Year	Parameters			
		SO ₂	NO ₂	SO ₂ + NO ₂	PM ₁₀
Jamshedpur	2010	35	48	83	153
	2011	36	48	84	152
New Delhi	2010	5	55	60	261
	2011	6	61	67	222
Kolkata	2010	11	62	73	99
	2011	12	65	77	113

Annual average concentration in μg/m³.

pinhole. Prior to analysis of samples, the instrument was calibrated by using pure Silicon at the peak of 522.28 cm⁻¹.

The third sets of experiments were performed by subjecting the exposed samples to electrochemical tests in 1% sodium chloride solution prepared by dissolving analytical reagent grade chemical in double distilled water. This test helped to assess the protective properties of the film formed on their surface during the period of their exposure at different sites. Anodic polarization studies were performed at scan rate of 0.1mV/sec. Although ASTM G59-97 (Reapproved 2003) recommends the scan rate of 0.6 V/h (0.16 mV/s), we preferred to keep it in lower side i.e. at 0.1 mV/s. This is due to the reason that a lower scan rate allows to generate a more stable current than the higher scan rate of the potential. The results are also more reliable and reproducible as reported by Zhang et al. (2009). The aim of the scan rate is to perturb the corroding interface with minimum capacitive effects at each point of the scan. A higher potentiodynamic scan rate will need too high current to hold the indicated potential. This has underestimating effect on the polarization resistance and shows higher corrosion current. In view of this a slower rate of scan is always desired. However the scan rate should not be so slow that it damages the specimen, especially during the anodic polarization. The experiments were performed in an electrochemical cell exposing the working electrode area of 2 cm². Two graphite rods at two sides of the working electrode were fitted, which acted as auxiliary electrodes. The reference electrode was a saturated calomel electrode (SCE). A luggin capillary was used to provide electrolytic contact between the calomel electrode and electrochemical cell. Electrochemical experiments were conducted by using a Gamry Potentiostat (supplied by M/S Gamry Instruments of USA). The data were analysed using DC 105 (for DC polarization) and CMS 300 (for EIS) software supplied by the manufacturer of the instrument. All the tests were performed at the temperature 30±2 °C. The temperature of the test electrolyte was maintained by circulating controlled temperature water through Teflon coated copper tubes immersed in the test electrolyte.

3. Results

Corrosion rates of industrial grade copper exposed in the atmosphere of Jamshedpur, New Delhi and Kolkata for two years (2010 and 2011) and their % decrease after one year of exposure are shown in Table 2.

The data incorporated in the above table show that corrosion rate during the first year of exposure of the metal at New Delhi was the highest and at Kolkata it was the lowest. During the second year of exposure of the samples, a substantial decrease in the rate of corrosion at all the places took place. The most noticeable decrease in the rate of corrosion was for the samples exposed at Jamshedpur where 83% reduction took place. It was very interesting observation and needs further discussion. Copper metal is considered most

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