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Review Article

Study the Corrosion and Corrosion Protection of Brass Sculpture by Atmospheric Pollutants in Winter Season

Rajesh Kumar Singh* and Noor Alam

Department of Chemistry, J P University, India

***Corresponding author:** Rajesh Kumar Singh, Department of Chemistry, J P University, India

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Abstract

Brass is an important metalloid which is used in construction of sculptures. It is noticed that sculpture of brass is corroding due to interaction of pollutants. The pollutants develop chemical and electrochemical reaction on the surface of base material. Their concentrations of corrosive pollutants are increased in winter season. The air quality becomes very poor in winter season. Inside sculpture different forms of corrosion are observed like galvanic, pitting, stress, crevice etc. The major components of pollutants are oxides of carbon, oxides of nitrogen, oxides of sulphur, ammonia, ozone and particulates. Among these pollutants oxides of sulphur and ammonia are major corroder of brass. Ammonia is observed moist air to form ammonium hydroxide. It produces chemical reaction with brass metal and form complex compounds like $[Zn(NH_4)_4](OH)_2$, $[Zn(NH_4)_4]SO_4$, $[Zn(NH_4)]CO_3$, $[Cu(NH_4)_4](OH)_2$, $[Cu(NH_4)_4]SO_4$, $[Cu(NH_4)]CO_3$ etc. Oxides of sulphur react with moist air to exhibit sulphurous and sulphuric acids. They interact with brass to develop corrosion cell zinc metal and it is oxidized into Zn^{2+} ions and these ions are active to humidity and carbon dioxide to yield $Zn(OH)_2.ZnCO_3.2H_2O$. Copper is converted into Cu^{2+} and it reacts with moist air and carbon dioxide to produce $Cu(OH)_2.Cu(CO_3)_2$ and these complex compound detached on the surface of brass metal by rain water. These pollutants change their physical, chemical and mechanical properties and they also tarnish their facial appearance. Brass' sculpture is affected by uniform corrosion. This type of corrosion can be control by nanocoating and electrospray techniques. For this work (6Z)-5,8-dihydrazono-5,8-dibenzo[a,c][8]annulene and TiO2 are used as nanocoating and electrospray materials. The corrosion rate of material was determined by gravimetric and potentiostat technique. The nanocoating and electrospray compounds are formed a composite layer on surface of base metal. The formation of composite layer is analyzed by thermal parameters like activation energy, heat of adsorption, free energy, enthalpy and entropy. These thermal parameters were calculated by Arrhenius, Langmuir isotherm and transition state equations. Thermal parameters results are depicted that both materials are adhered with sculpture through chemical bonding. The surface coverage area and coating efficiency indicates that nanocoating and electrospray are produced a protective barrier in ammonia and sulphur dioxide atmosphere.

Keywords: Brass sculpture; Corrosion; Atmospheric pollutants; Nanocoating; Electrospray; Sulphur dioxide; Composite barrier

Introduction

The sculpture of brass comes in contact of contaminated air thus its deterioration starts for protection various types methods can be applied [1]. Brass [2] has major components is copper and zinc. Zn reacts the hot air to produce ZnO which is active in humidity [3] to convert into $Zn(OH)_2$. In moist air [4], they form CuO, ZnO, $Cu(OH)_2$ and $Zn(OH)_2$. Both metals are active with sulphur to yield Cu2S, CuS and ZnS and these metallic sulphides [5] react with moist air to give $Cu(OH)_2$, $Zn(OH)_2$, $CuSO_4$ and $ZnSO_4$. The hydroxides of these metals interact with CO_2 to produce $CuCO_3$ and $ZnCO_3$. Sulphur dioxide [6] is a culprit of brass. It undergoes with $Cu(OH)_2$ and $Zn(OH)_2$ to convert into $CuSO_4$ and $ZnSO_4$. Moist SO_2 yields H_2SO_3 and H_2SO_4 whereas they create acidic environment [7] for brass and generate corrosion cell on their surface. It accelerates disintegration [8] in metal components of sculpture of brass. Brass is highly sensitive to ambient of ammonia gas [9]. It interacts with humid atmosphere [10] to NH4OH and it deposits on the surface brass metal [11] thus it converts into a complex layer of $[Cu(NH_3)_4](OH)_2$ and $[Zn(NH_3)_4](OH)_2$ that layer erosion starts in rain water. $[Cu(NH_3)_4](OH)_2$ and $[Zn(NH_3)_4](OH)_2$ complex compounds [12] come in contact of H2SO4 environment to produce $[Cu(NH_3)_4]SO_4$

and $[Zn(NH_3)_4]SO_4$ that complex layer is eroded in rain water. In acidic medium brass outer face has developed $CuSO_4$ and $ZnSO_4$ when dust particulates [13] are deposited on their surface which contains Fe to remove Cu and Zn from outer surface. Dust particulates are possessed oxides of alkali metal in presence of moisture, it produces NaOH or KOH [14] that is create hostile environment for Zn and it forms complex compound [15] $Na_2[Zn(OH)_4]$or $Na[Zn(OH)_3.H_2O]$ or $Na[Zn(OH)_3.(H_2O)_3]$. The oxides of NO_2 reacts with moist air to give HNO_3 that acid produces chemical reaction with Cu and it converted into $Cu(NO_3)_2$. Some organic acids [16] available in air like acetic acid which develop corrosive environment for Cu and Zn which converts Cu into $Cu_2(CH_3COO)_4.H_2O$ and Zn into $(CH_3COO)_6.Zn_4O$ complex compounds [17]. They are eroded by rain water on the surface of brass. Organic compounds [18] like amnio and sulpur increased day by day in atmosphere. They develop hostile environment for brass and corroding it. Corrosive pollutants [19] concentrations like oxides of carbon, oxides of nitrogen, oxides of sulphur, hydride of sulphur and nitrogen, ozone and particulates are enhanced due to industrials wastes, effluents, flues and other factors are like burning of coals, woods and cow dung cakes. Harmful pollutants [20] come into atmosphere through agricultural wastes, human wastes, pharmaceutical wastes, household wastes, food wastes and decomposition of living things. Various types of transports like road, water and air are evolving CO, NO_2 and SO_2 gases which produce acidic environments for brass. Several types of techniques are used to control the corrosion of brass like metallic coating; polymeric coating, paint coating, organic and inorganic coating of materials but these didn't give satisfactory results in corrosive medium. Some organic and inorganic inhibitors are applied to protect the corrosion of materials in acidic but they provide good results. Hot dipping, electroplating and galvanization techniques is used as protective tools for brass corrosion in acidic medium but these methods don't shave base metals. In this work it is to mitigate corrosion of brass corrosion by nanocoating and filler techniques. These materials form composite barrier on the surface base metal and blocked porosities and stop diffusion or osmosis process of pollutants.

Experimental

Brass coupons 15sqcm were taken for experimental analysis. Samples surface were rubbed with emery paper, rinsed with acetone, dry them and kept into desiccators. Sample kept 20meter height of roof in open sky and it observed that colour of brass can be changed. Corrosion rate was determined in winter season by weight loss method. The concentration of SO_2 in November 75ppm, December 90ppm, January 105ppm and February 120ppm and temperatures recorded in this period were 298K, 294K, 291K and 295K. Synthesis organic compound (6Z)-5,8-dihydrazono-5,8-dibenzo[a,c][8]annulene used as nanocoating and TiO_2 as filler and corrosion of brass metal calculated in above mentioned concentrations and temperatures in winter season. Both compounds formed a composite barrier on surface of base metal (Figures 1-4). Surface adsorption phenomenon studied by thermal parameters like activation energy, heat of adsorption, free energy, enthalpy and entropy.Potentiostat/Galvanostat model EG&G used for corrosion potential, corrosion current and corrosion current density. Brass sample put between H2|Pt electrode as auxiliary electrode and $Hg_2Cl_2|HgCl_2$ electrode reference electrode.

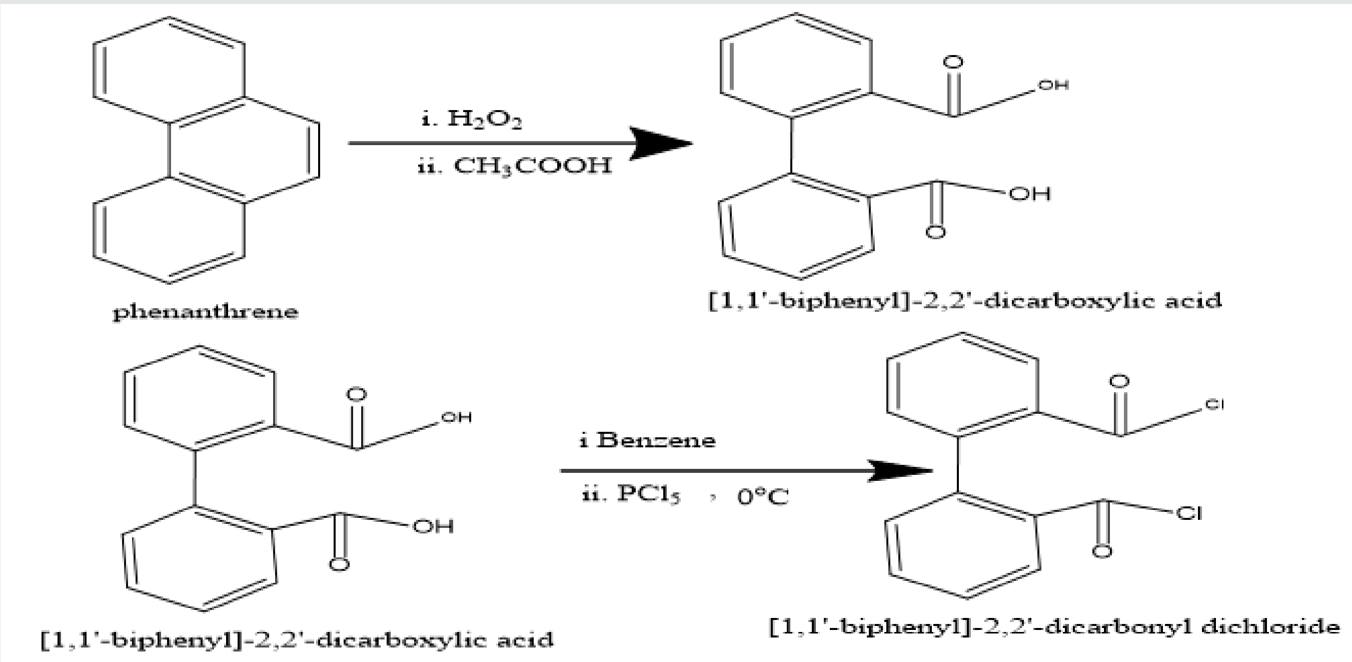

Figure 1

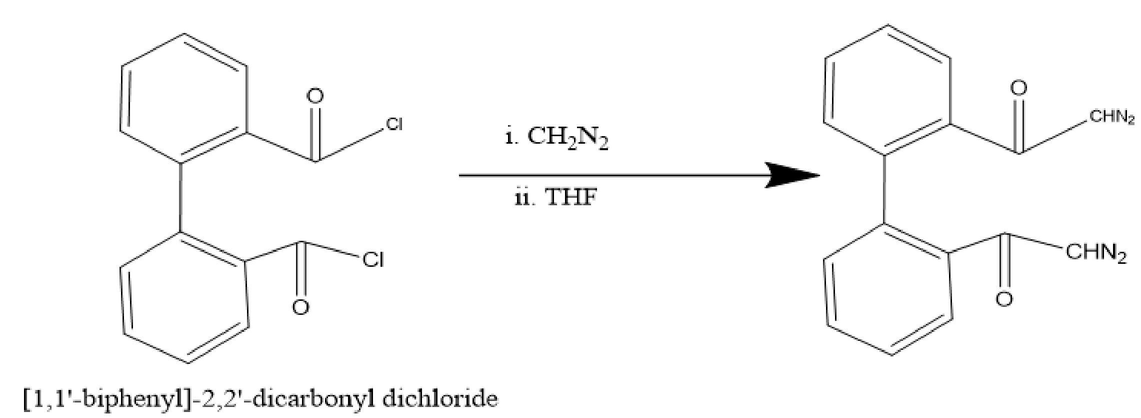

Figure 2

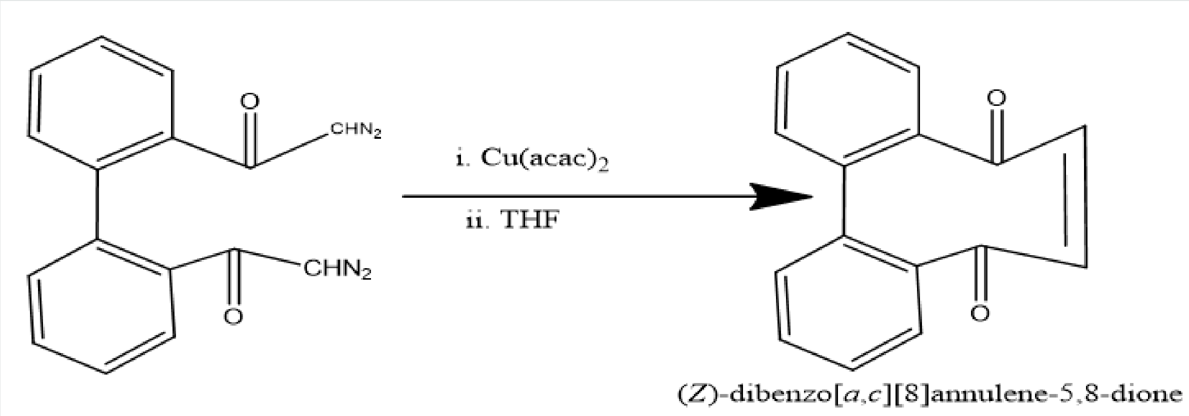

Figure 3

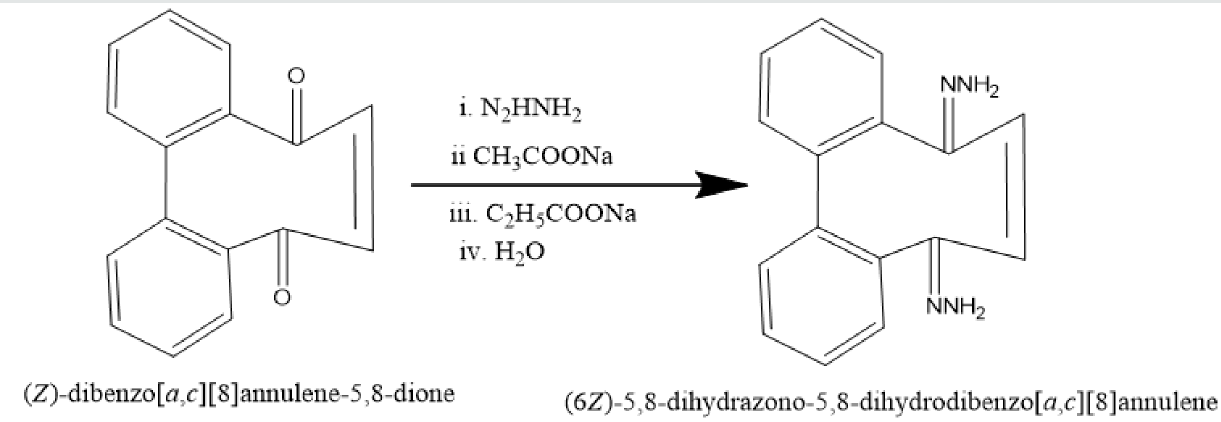

Figure 4

Synthesis of (6Z)-5,8-dihydrazono-5,8-dibenzo[a,c][8] annulene

Phenatharene was oxidized into [1,1'-biphenyl]-2,2'-dicarboxylic acid by the use of H_2O_2 in presence of CH_3COOH. When [1,1'-biphenyl]-2,2'-dicarboxylic acid was treated in PCl_5 in benzene solution at 0 °C temperature, [1,1'-biphenyi]-2,2'-dicarbonyl chloride was obtained. It reacted with diazomethane to produce yield [1,1'-biphenyl]-2,2'-dicarboxodiazomethan which heated $Cu(acac)_2$ in presence THF to yield (Z)-dibenzo[a,c][8]annulene-5,8-dione. It was used with hydrazine hydrate in ethyl alcohol to give (6Z)-5,8-dihydrazone-5,8-dihydrodibenzo[a,c][8]annulene.

Results and Discussion

Brass metal was exposed in moist SO_2 environment in 75ppm, 90ppm, 105ppm and 120ppm concentrations and 298 ^{0}K, 294 ^{0}K, 291 ^{0}K and 295 ^{0}K temperatures. The corrosion rate of brass metal was determined in winter season without coating and with coating (6Z)-5,8-dihydrazone-5,8-dihydrodibenzo[a,c][8]annulene and TiO_2 electrospray of by weight loss formula K= 534 W/DAT (where W is weight loss, D is density and T is time) and their values were mentioned in (Table 1).

Table 1: Corrosion of Brass Sculpture in Winter Season in SO_2 medium.

Month	Days	SO_2 (PPM)	K_o(mmpy)	$logK_o$			Temp(^{0}K)	(1000X1/T)K^{-1}
Nov	30	75	3456	3.538			298	3.36
Dec	31	90	4073	3.609			294	3.40
Jan	31	105	4356	3.639			291	3.54
Feb	28	120	4448	3.648			295	3.38
Corrosion Control of Brass Sculpture by nanocoating of (6Z)-5,8-dihydrazone-5,8-dibenzo[a,c][8]annulene [NC] in SO_2 Medium								
Month	**Days**	**SO_2 (PPM)**	**C(mM)**	**K(mmpy)**	**logK**	**log(θ/1-θ)**	**Temp(K)**	**(1000X1/T)K^{-1}**
Nov	30	75	25	1723	3.236	-0.281	298	3.36
Dec	31	90	30	1975	3.295	-0.513	294	3.40
Jan	31	105	40	2204	3.343	-0.495	291	3.54
Feb	28	120	45	2086	3.319	-0.545	295	3.38
Corrosion Control of Brass Sculpture by Electrospray of TiO_2 in SO_2 Medium								
Month	**Days**	**SO_2 (PPM)**	**C (mM)**	**K(mmpy)**	**logK**	**log(θ/1-θ)**	**Temp(K)**	**(1000X1/T)K^{-1}**
Nov	30	75	5	1280	3.107	0.393	25	3.36
Dec	31	90	10	1604	3.205	0.234	21	3.40
Jan	31	105	15	1128	3.052	0.416	18	3.54
Feb	28	120	20	1099	3.041	0.331	22	3.38
Surface coverage area and % Coating efficiency by Nanocoating material (6Z)-5,8-dihydrazono-5,8-dibenzo[a,c][8]annulene [NC] in SO_2 Medium								
Month	**Days**	**SO_2 (PPM)**	**C(mM)**	**θ**	**% CE**	**log(θ/1-θ)**	**Temp(K)**	**(1000X1/T)K^{-1}**
Nov	30	75	25	0.5014	50.14	-0.281	298	3.36
Dec	31	90	30	0.5150	51.50	-0.513	294	3.40
Jan	31	105	40	0.4940	49.40	-0.495	291	3.54
Feb	28	120	45	0.5310	53.10	-0.545	295	3.38
Surface coverage area and % Coating efficiency by Electrospray of TiO_2 in SO_2 Medium								
Month	**Days**	**SO_2 (PPM)**	**C(mM)**	**θ**	**% CE**	**log(θ/1-θ)**	**Temp(K)**	**(1000X1/T)K^{-1}**
Nov	30	75	5	0.6296	62.96	0.393	298	3.36
Dec	31	90	10	0.6061	60.61	0.234	294	3.40
Jan	31	105	15	0.7410	74.10	0.416	291	3.54
Feb	28	120	20	0.7529	75.29	0.331	295	3.38

The corrosion rate of brass metal was recorded in the months of November, December, January and February, the results (Table 1) was shown that corrosion rate of metal increased in January to February but theses values were reduced with coating and filler materials like (6Z)-5,8-dihydrazone-5,8-dihydrodibenzo[a,c][8] annulene and TiO_2. It was clearly noticed in (Figure 5) K versus Month. Brass metal kept into 75ppm, 90ppm, 105ppm and 120ppm of SO_2 medium in month of Nov, Dec, Jan and Feb without coating. It was coated with 25mM, 30mM, 40mM and 45mM concentrations of (6Z)-5,8-dihydrazone-5,8-dibenzo[a,c][8]annulene, and again kept into same concentrations of SO_2. After coating of (6Z)-5, 8-dihydrazone-5,8-dibenzo[a,c][8]annulene electrospray coating of TiO_2 used at 5mM, 10mM, 15mm and 20mM concentrations and same concentrations SO_2 Nov to Feb. The corrosion rates of in these three cases were written in (Table 1). These results were shown that corrosion rates without coating increased, it values decreased coating with (6Z)-5, 8-dihydrazone-5,8-dibenzo[a,c][8]annulene but their values more reduced with TiO_2 electrospray. These trends were shown in (Figure 6) which plotted K versus C. The corrosion rates of brass metal at different temperatures 298 ^{0}K, 294 ^{0}K, 291 ^{0}K and 295 0K without and with coating were recorded in (Table 1). The addition of nanocoating and electrospray were reduced the corrosion rates as temperatures variation, it noticed in K versus T in (Figure 7).

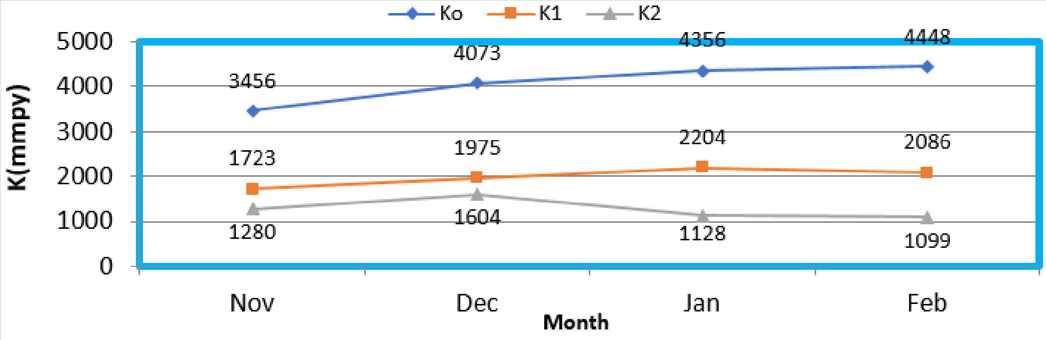

Figure 5: K(mmpy) V_s Months for brass metals.

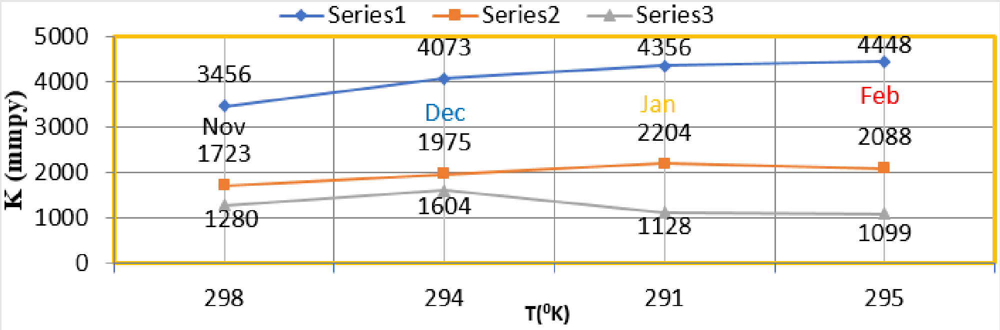

Figure 6: KV_s T nanocoating and electrospray.

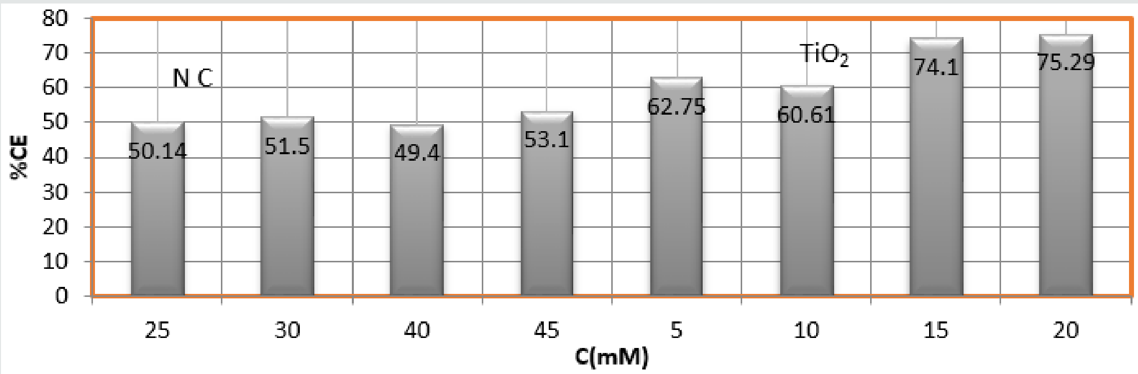

Figure 7: %CE V_s C(mM) nanocoating and electrospray.

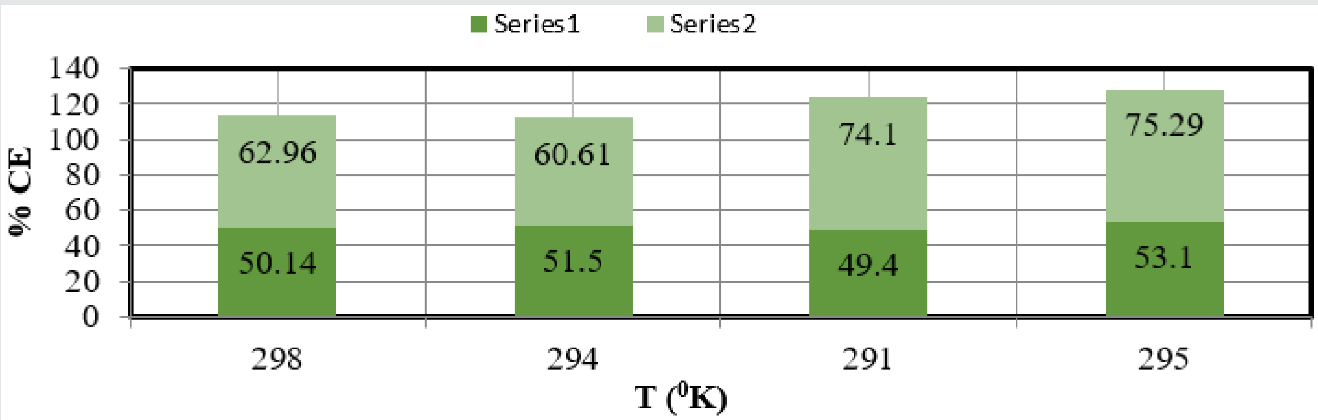

Figure 8: %CE V_s T for nanocoating and electrospray.

(Figure 8) plot between %C (percentage coating efficiency) versus C (concentrations in mM) indicated that nanocoating compound (6Z)-5, 8-dihydrazone-5,8-dibenzo[a,c][8]annulene increased coating efficiency but TiO_2 electrospray produced more coating efficiency with respect of nanocoating compound. The values of % coating efficiency were calculated by formula %CE = (1-K/Ko) X100 (where Ko is corrosion rate without coating and K is corrosion rate with coating) and their values were given (Table 1). (Figure 9) show plot between %C (percentage coating efficiency) versus T (temperature in K). This figure indicated that percentage coating efficiency enhanced as temperatures varies in Nov to Feb months and their values were recorded in (Table 1). Figure 6 plotted between θ (surface coverage area) versus C (concentration in mM) and covered areas were produced by (6Z)-5, 8-dihydrazone-5,8-dibenzo[a,c][8]annulene and TiO_2 were mentioned in (Table 1). The results were shown that nanocoating compound occupied less surface areas with respect of electrospray. The surface coverage area developed by nanocoating and electrospray compound was calculated by formula θ = (1- K/Ko). (Figure 10) plotted between θ (surface coverage area) versus T (temperature) noticed that temperatures were varies from Nov to Dec but surface coverage area and electrospray values were increased and their values were written in (Table 1).

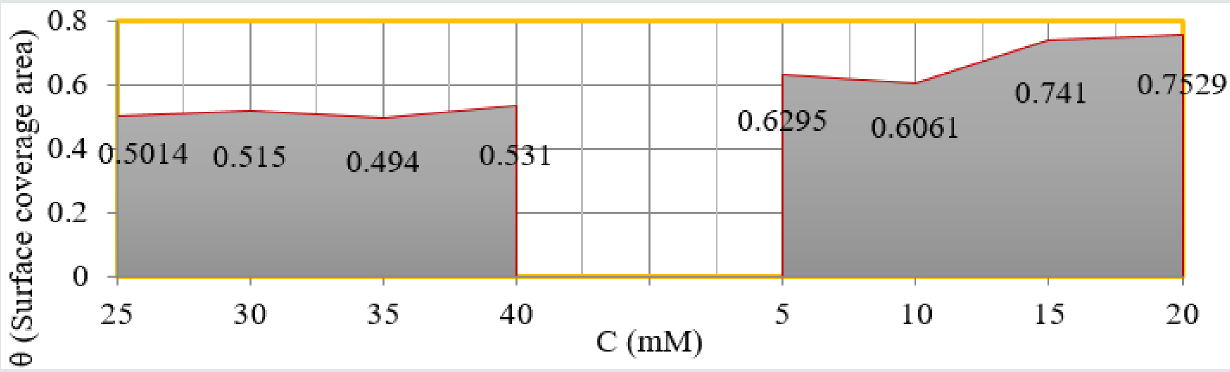

Figure 9: V_s C nanocoating and electrospray.

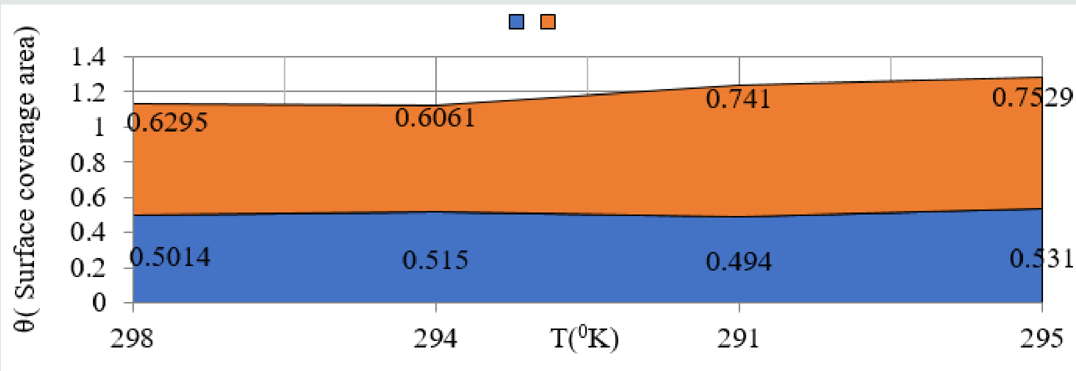

Figure 10: θ V_s T for nanocoating and electrospray.

Table 2: Thermal Parameters of Brass Sculpture in Winter Season by Nanocoating of (6Z)-5,8-dihydrazono-5,8-dibenzo[a,c][8] annulene [NC] in SO_2 Medium.

Month	C(mM)	Temp(K)	SO_2 PPM	Ea_o (kJ/mol)	Ea(kJ/Mol)	q (kJ/mol)	ΔG (kJ/Mol)	ΔH (kJ/mol)	ΔS(kJ/K)
Nov	25	298	75	234	222	-18.06	-321	-188	-151
Dec	30	294	90	236	229	-33.23	-328	-194	-153
Jan	40	291	105	246	236	-35.52	-339	-199	-151
Feb	45	295	120	228	221	-35.23	-320	-187	-150

Composite surface formation was studied by Arrhenius equation, Langmuir isotherm and others thermal parameters like activation energy, heat of adsorption, free energy, enthalpy and entropy and their values were recorded in (Table 2). (Table 2) Thermal Parameters of Brass Sculpture in Winter Season by Nanocoating of (6Z)-5,8-dihydrazono-5,8-dibenzo[a,c][8]annulene [NC] in SO_2 Medium. Activation energy of without coating, with coating and electrospray coating were determined by Arrhenius equation d(logK)/dT = A – Ea/2.303RT and their values were recorded in (Table 2). The plot between logK versus 1/T was found to be straight line as shown in (Figure 11). The plot between log K and 1/T found to be straight line. It observed that activation before coating activation energy high but decreased after coating. These trends indicated that nanocoating compound adhered on the surface of base metal. Heat of adsorption was calculated by Langmuir isotherm log(θ/1-θ) = log(AC) – q/2.303R T and their values were mentioned in (Table 2). Its values were found to negative, it indicated nanocoating compound formed chemical bond with base metal. (Figure 12) log(θ/1-θ) versus 1/T proved results of heat of adsorption.Free energy values of nancoating compound were determined by formula ΔG = -2.303 RT log(33.3K) and their values were recorded in (Table 2). Their values found to be negative; it noticed that nanocoating compound adhered on the surface of base metal by chemical bond. Enthalpy and entropy values of nanocoating and electrospray compounds were calculated by transition state equation K=k T/N h $e^{\Delta S/R}$ $e^{-\Delta E/RT}$ and their values were mentioned in (Table 2). These values were found to be negative which indicated these compounds adhered on the surface of metals. All thermal parameters versus T (temperature) plotted in (Figure 13) which indicated composite barrier formed on surface of base metal. Thermal parameters Values of TiO_2 eleectrospray activation energy, heat of adsorption, free energy, enthalpy and entropy were written in (Table 3) and their plot against T (temperature) in (Figure 14). (Table 3) results indicated electrospray compound formed chemical bond with nanocoating compound. (Table 3) Thermal Parameters of Brass Sculpture in Winter Season by Electrospray of TiO_2 in SO_2 Medium Potentiostat results were determined with help of equation I = βa βc/2.3 (βa+βc) Ic and corrosion rate K=0.128 X Ic X(E/d) (Ic is corrosion current, equivalent weight and d is density) and their values were written in (Table 4). (Figure 15) was plotted ΔE(corrosion potential versus I(corrosion current density). The results of (Table 4) observed that without coating corrosion potential high but with coating nanocoating and electrospray reduced corrosion potential. (Table 4) Potentiostat results in SO_2 in meduim with nanocoating and electrospray.

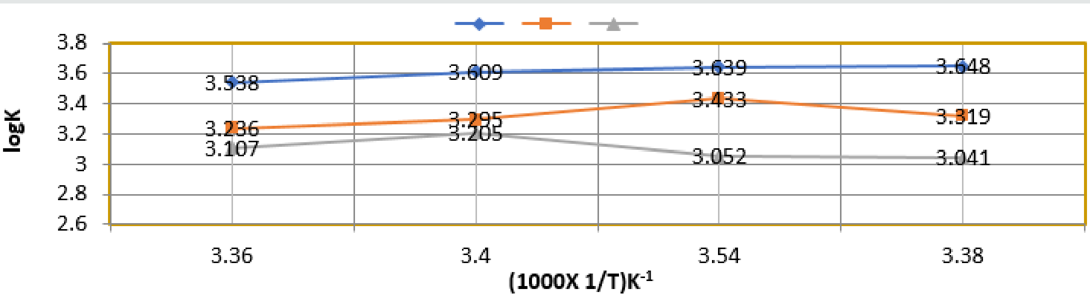

Figure 11: logK V_s 1/T nanocoating and electrospray.

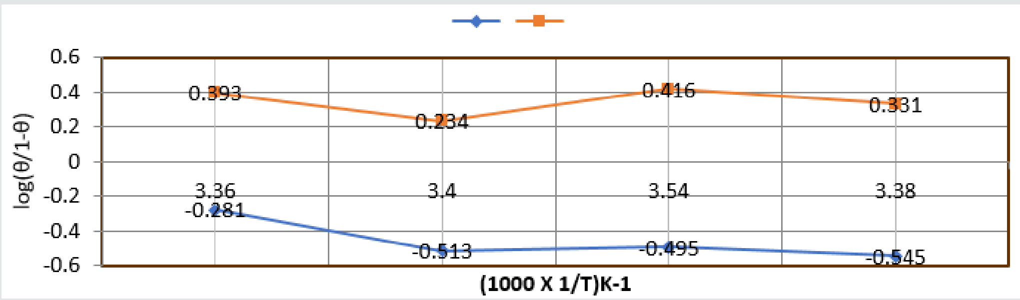

Figure 12: log(θ/1-θ) V_s 1/T nanocoating and electrospray.

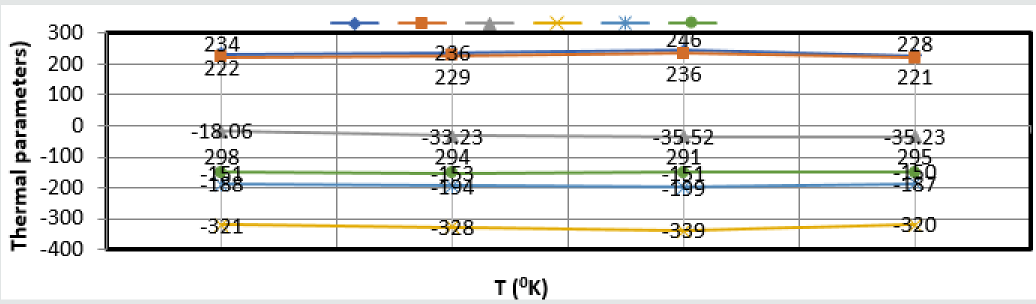

Figure 13: Thermal parameters V_s T for nanocoating and electrospray.

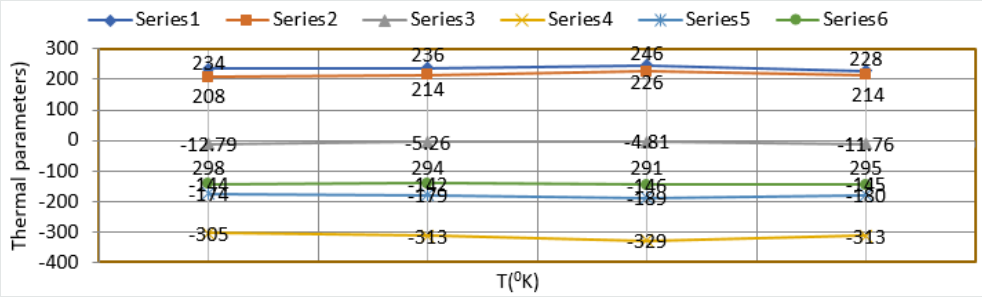

Figure 14: Thermal parameters V_s T nanocoating and electrospray.

Table 3: Thermal Parameters of Brass Sculpture in Winter Season by Electrospray of TiO_2 in SO_2 Medium.

Month	C(mM)	Temp(K)	SO_2 PPM	Ea_o	Ea	q	ΔG	ΔH	ΔS
Nov	5	298	75	234	208	-12.79	-305	-174	-144
Dec	10	294	90	236	214	-05.26	-313	-179	-142
Jan	15	291	105	246	226	-04.81	-329	-189	-146
Feb	20	295	120	228	214	-11.76	-313	-180	-145

Table 4: Potentiostat results in SO_2 in meduim with nanocoating and electrospray.

ΔE	I	βa	Ba	Ic	logIc	C(PPM)	Months	CN(mM)	CS(mM)	K
540	180	230	151	51	1.71	75	Nov	00	00	3457
545	195	257	147	67	1.82	90	Dec	00	00	3578
559	205	265	187	78	1.89	105	Jan	00	00	3798
578	234	278	199	85	1.97	120	Feb	00	00	3876
Potentiostat results in SO_2 in medium with nanocoating of (6Z)-5,8-dihydrazono-5,8-dibenzo[a,c][8]annulene [NC]										
451	175	180	198	45	1.65	75	Nov	25	00	1567
421	163	175	215	57	1.75	90	Dec	30	00	1610
396	154	165	231	65	1.81	105	Jan	40	00	1701
334	145	154	252	71	1.85	120	Feb	45	00	1802
Potentiostat results in SO_2 in medium with Electrospray of TiO_2										
401	161	147	260	41	1.61	75	Nov	00	5	1378
376	144	140	265	49	1.69	105	Dec	00	10	1402
347	137	138	271	58	1.76	105	Jan	00	15	1512
231	131	128	278	63	1.79	120	Feb	00	20	1598

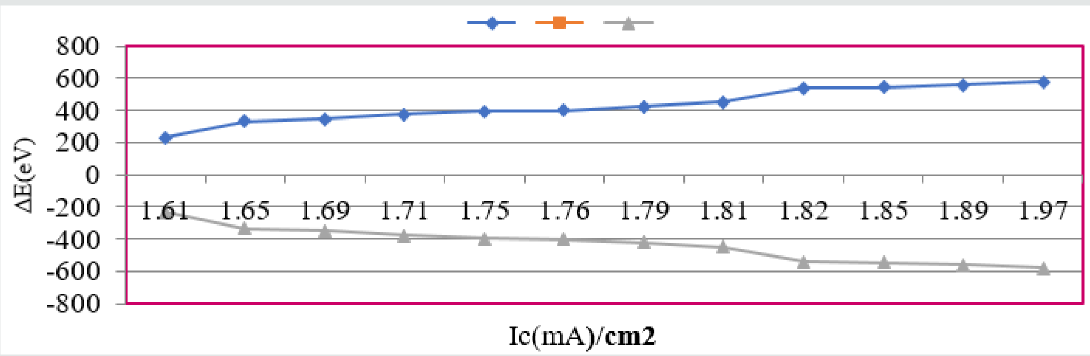

Figure 15: ΔE V_s Ic(mA) for nanocoating and electrospray.

Conclusion

It observed that winter season SO_2 concentration increased. In this season humidity level found to more so it oxidized sulphur dioxide into sulphuric acid and created hostile environment for brass sculpture. It corroded zinc into zinc sulphate in longer period it produced leaching corrosion. Such types of corrosion controlled by the use of nanocoating of (6Z)-5,8-dihydrazono-5,8-dibenzo[a,c][8]annulene and TiO_2 electrospray. The results of activation energy, heat of adsorption, free energy, enthalpy and entropy values indicated that nanocoating compound adhered with chemical bonding. Thermal parameters results of electrospray confirmed that TiO_2 bonded with (6Z)-5,8-dihydrazono-5,8-dibenzo[a,c][8] annulene chemical bonding. Both compound created composite barrier on the surface of base metal which produced anticorrosive barrier. The nanocoating compound developed lot of porosities during coating. These porosities blocked by electrospray and it increased coating efficiency and surface coverage area.

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