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Corrosion Behaviour of High Phosphorus Containing Cu-Cr Weather Resistant Steel

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Abstract

Three steels viz., G22(Fe-0.072C-0.028 Si-0.20Mn-0.19P-0.38Cr-0.24Cu-0.17Ni), G32(Fe-0.072C- 0.061 Si-0.23 Mn-0.19P-0.64Cr-0.14Cu) and SCOR (Fe-0.09C-0.37Si-0.42Mn-0.15P-0.37Cr-0.35Cu-0.31Ni) were made through ingot metallurgy route and hot rolled to 3 mm thick sheets. Tafel extrapolation study of freshly ground samples in 3.5% NaCl solution revealed similar corrosion rates for all steels. Another technique employed to evaluate the effect of phosphorus on the formation of protective rust layer was electrochemical impedance spectroscopy (EIS) test after exposing the samples in an atmospheric exposure rack for four months on the roof top of a fourth floor building. Results revealed that the polarization resistance or rust resistance of G22 and SCOR steels was comparable and similar to that of a reference steel SAILCOR (Fe-0.07C-0.48 Si-0.31Mn-0.118P-0.41Cr-0.32Cu-0.21Ni), which is produced at Bokaro Steel plant. The role of phosphorus in improving atmospheric corrosion resistance was understood from this study.

Keywords: Tensile properties; Impact energy; Hot ductility; EIS; Impedance

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Introduction

A significant amount of economic loss occurs due to premature failure of engineering components and structures in industry and infrastructures because of degradation by corrosion. The loss due to corrosion is quite high even in developed countries to the extent of 5% of gross domestic product (1).Atmospheric corrosion is more severe in the chloride and sulphur dioxide laden environments (2, 3). The use of weathering steel instead of carbon steel, having less than 3 wt.% alloying elements of Cr, Ni, Cu, and P, has been most effective and economical way of mitigating atmospheric corrosion (4-6). These weather resistant steel provide optimum balance of corrosion resistance and strength due to the presences of various alloying elements. A compact rust layer is formed during long-term exposure to the atmosphere. This compact rust layer gradually becomes denser over time and eventually protects the steel from corrosive environment & thus corrosion is inhibited by the corrosion product. Minor damage to the rust heals itself; therefore maintenance is greatly reduced. In 1933, the first commercial weather resistant steel of this type was introduced in the market with the trade name of USS-CORTEN (7). Phosphorus has been one of the crucial elements in weathering steels owing to the reported catalytic effect on the formation of amorphous type FeOOH compact rust layer (8-10). The effect of phosphorus is more pronounced in decreasing corrosion rate of steel, especially, during long term exposure (11, 12).

Some of the commercial weather resistant steels containing relatively higher amounts of phosphorus than that of conventional steels are SAILCOR grade steel produced by Bokaro Steel Plant, SAIL, India and patented CORTEN steel, USA. The range of chemical composition (in wt.%) of the commercial CORTEN steels is: C: ≤0.12-0.16; Si: 0.25-0.75, Mn: 0.20-1.2, Cr:0.40-1.25, Cu: 0.25-0.55, V:0.02-0.10, Ni: ≤0.65, P: ≤0.15 and that of SAILCOR is C: 0.12 max, Mn: 0.2-0.5, S: 0.04 max, Si: 0.25-0.75, Al: 0.08 max, Cu: 0.25-0.65, Ni: 0.17-1.50, Cr: 0.30-0.65, P: 0.145 max. However, the actual phosphorus concentration in the weathering grade steel is found to be around 0.10 wt.% (11-12) and that in industrial heats of SAILCOR grade steel is in the range of 0.1-0.12 wt.%. Both CORTEN and SAILCOR grade commercial steels can contain Ni as high as 0.65% and 0.50% respectively. As Ni is a costlier element, the reduction of Ni content in weathering steel without compromising its corrosion and mechanical properties will be appreciated. Due to lower melting temperature of Cu, addition of Ni (equal amount or less than that of Cu content) is beneficial for reducing hot-shortness of steel where Ni increases solubility of Cu in austenite (13). As Cu has been used for improving corrosion resistance properties of steel, phosphorus concentration can be increased to compensate reduction in Cu content.

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In this work, suitable alloy designs have been made keeping Cu and Ni content low with high phosphorus content. The corrosion resistance behaviours have been evaluated from the Tafel extrapolation method and electrochemical impedance spectroscopy study.

Materials and Experimantal Procedures

Laboratory Heat Making and Hot Rolling

Three numbers of heats of low alloy steels of 25 kg each were made utilizing a high frequency induction-melting furnace of 100 kg capacity. Steel scraps of C-Mn rail steel had been added to soft iron (0.001 wt.% C) to balance carbon content. In order to obtain Cr, P, Cu and Ni in the desired range, Fe-P and Fe-Cr ferro alloys along with Cu blocks and Ni lumps were added to the liquid steel in the furnace. Aluminium shots and wire were used for deoxidation purpose. The melts were cast into ingots of cross sectional dimensions 100 × 100 mm. Subsequently, the ingots were hot rolled in two stages. In the first stage, the ingots were rolled into 16 mm thickness plates through five passes after soaking at 1,150°C for 2.5 h. The finishing temperature was about 950°C, which is just above Ar^3 temperature. In the second stage, once again these plates were soaked for 30 min at 1,150°C and hot rolled into 5 mm thickness plate. The chemical composition of each steel was determined using an optical emission spectrometer and provided in Table 1. A commercial SAILCOR grade steel produced by Bokaro Steel Plant, SAIL, India and a laboratory made low carbon steel (LC) were used as references for comparison purpose.

Table 1. Chemical composition of steels (in wt.%).

Heats	C	Si	Mn	S	P	Cr	Cu	Ni	Al
LC	0.086	0.110	0.290	0.010	-	-	-	-	0.030
SCOR	0.090	0.370	0.420	0.010	0.150	0.370	0.350	0.310	0.030
G22	0.072	0.028	0.200	0.007	0.190	0.380	0.240	0.170	0.030
SAILCOR	0.070	0.480	0.310	0.008	0.118	0.410	0.320	0.210	0.012

Tafel Polarization

Samples for Tafel extrapolation study were ground successively to 1,200 grit SiC abrasive paper and were degreased with acetone. The polarization experiments were conducted in a standard flat cell (Princeton Applied Research, Ametek, USA) in the potential range –250 mV to +250 mV versus open circuit potential of samples, using a computer controlled potentiostat (Princeton Applied Research, Ametek-273A, USA). Polarization experiments were carried out in 3.5% NaCl solution of pH 6.8 for at least three numbers of samples for obtaining reproducibility of the results. It is reported that dissolved oxygen content is maximum at 3.5% NaCl and it will decrease either on decreasing or increasing from 3.5%. Furthermore, 3.5% NaCl was used to simulate the immersed corrosion in saline environment. A saturated silver-silver chloride (SSC) reference electrode with platinum counter electrode was used. The scan rate employed for the polarization studies was 0.25 mVs^{-1}. The corrosion current density (icorr) and zero current potential (Ecorr) were evaluated from Tafel polarization plots by Tafel extrapolation method as per ASTM Standard G3-89 [14, 15]. Corrosion rate in penetration units, mils/year (mpy) was calculated from icorr using the following equation [15]

$$\text{mpy} = i_{\text{corr}} \times \Lambda \times \frac{1}{\rho} \times \varepsilon \qquad (1)$$

where

Λ = 1.2866×10^5 [equivalents.sec.mil]/[Coulombs.cm.years]
i_{corr} = the corrosion current density in $Amps/cm^2$ (Amp = 1 Coulomb/sec)
ρ = density (7.86 $grams/cm^3$ for iron)
ε = equivalent weight (27.56 grams/equivalent, for iron)

Atmospheric Exposure Study

Test coupons were subjected to atmospheric exposure on the roof of the RDCIS Lab building at Ranchi, as per ASTM G 50-76 specification (Figure 1) to assess corrosion resistance in the form of electrochemical impedance of the rust layer. Three numbers of test coupons were exposed for each steel. The atmospheric exposure stand confirming to the above specification was prepared for this study. Electrochemical impedance spectroscopy (EIS) test was conducted in 3.5% NaCl solution of pH 6.8 after atmospheric exposure of 4 months (Oct'12- Feb'13) duration. Experiments were carried out for three numbers of samples for obtaining reproducibility of the results. EIS scan was carried out by applying a sinusoidal potential perturbation of 10 mV at the open circuit potentials with frequency sweep from 100 kHz to 100 mHz. All the experimental EIS data were obtained in VERSASTUDIO-2.03 software and modelled using Zsimpwin (version 3.21, Princeton Applied Research, Ametek, USA) software.

Figure 1. Atmospheric exposure racks with samples exposed in it.

Results and Discussion

Alloy Design and microstructure

The primary purpose of alloy design was to make the steel cost effective by decreasing costlier Ni. As provided in Table 1, the steel chemistries were designed to keep carbon content low (0.06-0.09%) and phosphorus content high (0.13-0.20%) in contrast to the CORTEN steel, where both carbon and phosphorus content are kept about 0.12% (7). Numbers of researchers have reported that phosphorus increase ductile to brittle transition temperature (DBTT). However, lower amount of carbon is beneficial in suppressing deleterious effect of phosphorus by replacing it from the grain boundaries (16, 17). Therefore, a combination of low carbon and high phosphorus steel chemistries were choosen in this work. Mesquitaa and H. J. Kestenbach (18) have investigated that on increasing silicon content toughness decreases due to the formation of coarser alloy carbides. Therefore, Si content of relatively high phosphorus steel G22 and G32 (0.19% P) is 0.028% and 0.061%, respectively. As phosphorus is a strong solid solution strengthening element, Mn content of these steel was decreased to 0.20 to 0.23% in contrast to 0.42% Mn in SCOR steel. G22 steel contains low Cu (0.24%) as it contains higher phosphorus than that of the SCOR steel. Due to lower melting temperature (1,085°C) of Cu, addition of equal amount of Ni or less than that of Cu content is beneficial for reducing hot-shortness of steel where Ni increases solubility of Cu in austenite (19). Therefore, as Cu content was lower in G22 steel than that in SCOR steel, Ni content was reduced from 0.31 to 0.17%. A low carbon steel (LC) and an industrially produced low alloy steel (SAILCOR) were used for comparative study. On comparing the percentage of alloying elements in SCOR, G22 and SAILCOR, it can be easily estimated that the cost of G22 steel will be cheaper as it has lower amounts of Ni and Cu in addition to Mn and Si.

Figure 2 shows the light optical photomicrograph of SAILCOR, SCOR, G22 and LC steels along cross section. All photomicrographs reveal ferrite (lighter phase) pearlite (darker phase) microstructure, which is typically observed in low carbon and low alloy steels

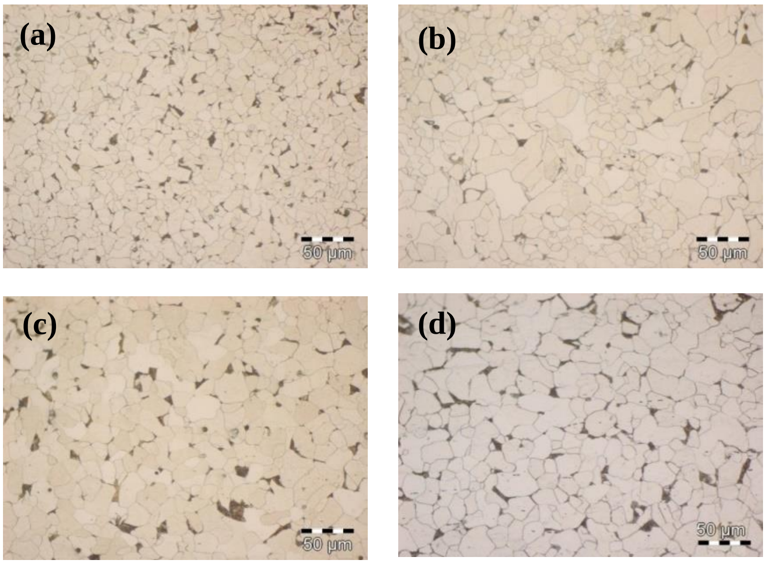

Figure 2. Optical photomicrograph of (a) SAILCOR, (b) G22, and (c) SCOR and (d) LC steel.

Tafel polarization

The obtained Tafel plots of LC, G22, SCOR and SAILCOR samples are shown in Figure 3. The cathodic branch of all Tafel plots exhibit diffusion controlled reaction as corrosion current remains nearly constant. The NaCl solution used for the potentiostatic Tafel polarization is aerated and neutral where the cathodic reaction consists of the oxygen reduction eduction ($O_2 + 2H_2O + 4e \rightarrow 4OH^-$) (20, 21). This is also evident from Pourbaix diagram of Fe-H_2O System (22). Therefore, as the diffusion controlled oxygen reduction is the only dominant cathodic reaction, the linearity of cathodic Tafel region is absent. The i_{corr} and corrosion rate (in mpy) obtained from the Tafel polarisation curves of samples under investigation are tabulated in Table 2. The E_{corr} of the Balasubramaniam (23), the corrosion rate of low alloy, C-Mn, and high phosphorus steel obtained from the Tafel extrapolation method in 3.5% NaCl between 6 to 10 mpy. The similar results have been also reported by other researchers.

As reported by Hudson (26), the corrosion rate obtained from weight loss method through immersion test in sea water of different grades of steels, e.g., mild steel (0.2% C), copper bearing steel (0.48-0.53% Cu), Cr-Cu Steel (0.42-0.51% Cr and 0.41-0.56% Cu) were 6.09, 5.65 and 6.08 mpy, while that of Cr- Mo-Ni steel (3% Cr-0.5% Mo –0.1% Ni) and Cr-Ni Steel (1% Cr-3% Ni) were 1.09 and 1.6 mpy, respectively. The reported corrosion rate of plain carbon steel (AISI 1020) after one year immersion in quiet sea water is 15 mpy and then decreases to 5 samples lies between -510 mV SCE to -550 mV SCE. As mpy after 1000 days. The corrosion rate of pure provided in Table 2, the corrosion rate of all samples is almost similar and in good agreement with the literature findings (23-25). As reported by Sahoo and iron is 12 mpy as obtained by the Tafel extrapolation method after 24 hours of immersion in unstirred, air saturated 3.5% NaCl solution (28).

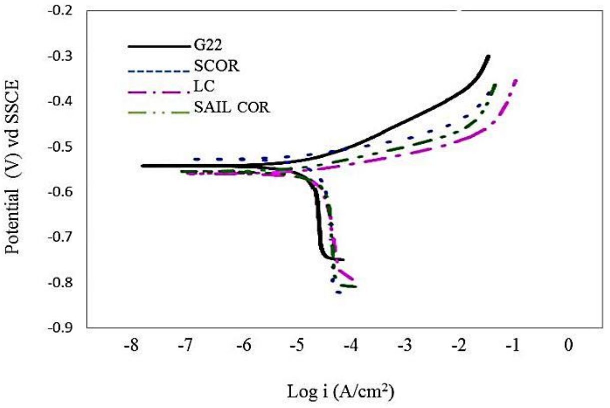

Figure 3. Tafel plots of obtained in 3.5% NaCl solution at scan rate 0.25 mV/s.

Table 2. Tafel parameters: E_{corr}, i_{corr} and corrosion rate (mpy) obtained by Tafel extrapolation method from Tafel plots.

Sample	E_{corr} (mV)	i_{corr} (A/cm^2)	Corrosion Rate (mpy)
G-22	527	14.48	6.64
SCOR	513	22.60	10
SAILCOR	541	20.56	9.42
LC	547	20.49	9.39

The random variation of corrosion rate between 6 to 10 mpy of low alloy and plain carbon steel could be assumed as a range which accommodate the error of sample preparation, the Tafel extrapolation method for calculating i_{corr} etc. In the present study, the corrosion rate of samples obtained by Tafel extrapolation method after 5 minutes immersion in 3.5% NaCl is in the range of 6.5 to 9.5 mpy. Thus, the Tafel polarization study showed that presence of alloying elements like Cu:0.30-0.35, Cr: 0.36-0.37, Ni: 0.20-0.31 and P: 0.10-0.20 have little effect on reducing corrosion rate in 3.5% NaCl as the corrosion rates of low carbon steel and low alloy steel are almost similar (Table 2). In order to understand the beneficial effect of the alloying elements, especially P, further studies on these steels had been carried out after allowing the samples to gather rust through standard atmospheric exposure test.

Atmospheric Exposure Test

In low alloy steel containing Cu, Ni, P and Cr, unlike immersion conditions, influence of mild or normal environment such as atmospheric exposure is beneficial in improving weathering properties of steel. It has been shown by Misawa et al. (29, 30) .that Cu, P and Cr in low alloy steel contributed to uniform dissolution of steel and facilitated uniform formation of protective rust composed of Fe_3O_4, α-FeOOH, α-FeOOH and γ- FeOOH on steel surface. For this reason, it was envisaged in this work to study the influence of atmospheric conditions on weathering behaviour of investigated steel after exposing the specimens on the roof top in atmospheric exposure racks. The variations of atmospheric temperature and relative humidity during the atmospheric exposure period (Oct'12-Feb'13) of samples are shown in Figure 4. The maximum average relative humidity is higher than 80%. It is well known that corrosion of metals and steel occurs during wet periods (31) .Furthermore, it is reported that wetness occurs when the relative humidity exceeds 80% (31) .Therefore, the exposure period was favourable to facilitate corrosion. The minimum temperature was recorded when percentage of relative humidity is maximum.

At the end of four month exposure, electrochemical impedance spectroscopy (EIS) of rusted samples was conducted in 3.5% NaCl solution and the results in the form of Nyquist plots are presented in Figure 5. It reveals that the impedance (Zreal) or the polarization resistance or rust resistance of the rust (maximum X-value of the semicircle in the high frequency region of plot) of SCOR, G22, and SAILCOR steels are comparable, while it is much higher (more than three times) than that of low carbon steel. Particularly, the semicircle portion of SCOR and G22 are similar, which imply equal Zreal value for both these steels. The Cu and Ni content in SCOR are 0.35% and 0.31% while that in G22 are 0.24% and 0.17%, respectively. On the other hand, P in SCOR is 0.15% and that in G22 is 0.19%. Therefore, the presence of higher amount of phosphorus in G22 steel contributed in improving weathering properties similar to that in SCOR steel containing higher amounts of Cu and Ni

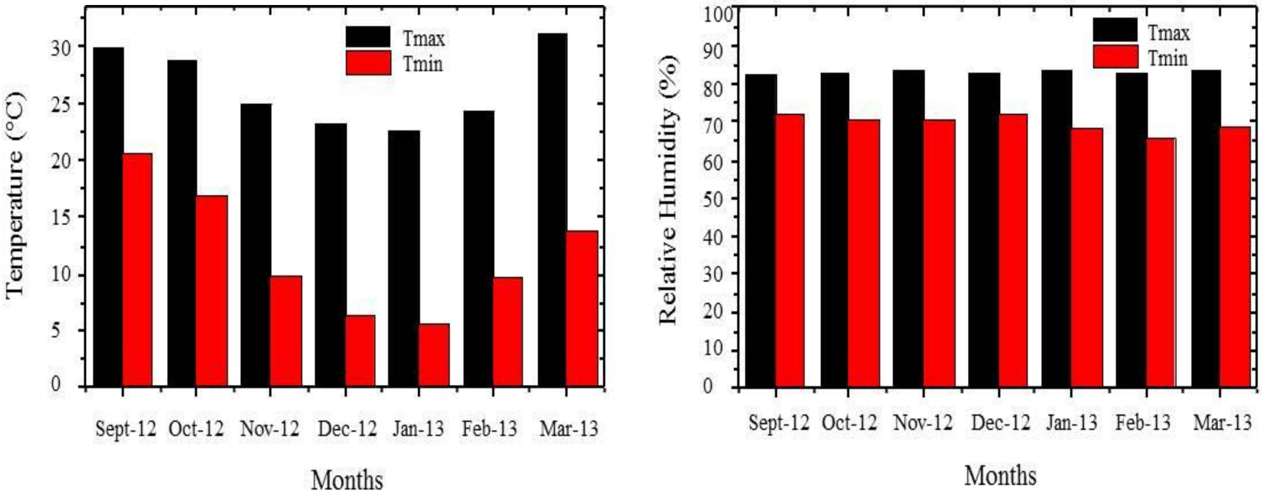

Figure 4. Variation of monthly average (a) temperature in degree centigrade and (b) percentage of relative humidity.

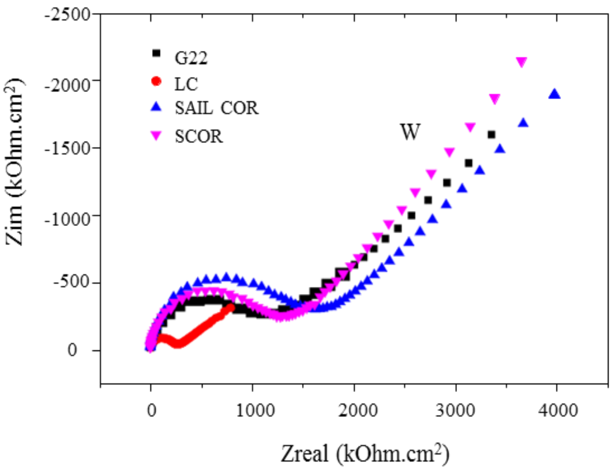

Figure 5. Nyquist plots of atmospheric exposure samples obtained in 3.5% NaCl solution.

The low frequency region of each Nyquist plot shows Warburg constant (W), which is usually at 45°angle to the real axis (32). The presence of Warburg constant has been attributed to the diffusion controlled electrochemical process involving diffusion of dissolved oxygen from the bulk solution to the electrode surface (32). As iron oxide/oxyhydroxide type rust has formed on all the samples, the Warburge diffusion is found in Nyquist plots.

Recent study of Sahoo et al. (33) has confirmed that addition of phosphate species to green rust changed dramatically the morphology as well as particle size of lepidocrocite from needle like cylindrical shape to fine globular shape. The finer particle size of rust will make it more compact and impervious. Therefore, the improved rust resistance of SAILCOR and developed steel have been understood. As the impedance of developed steels G22 and SCOR is nearly similar, the use of G22 steel for weathering purpose will be economical as the chemistry, especially alloying elements in SCOR are richer than that of G22.

Conclusions

1. Alloy chemistries with variations of phosphorus, copper and nickel were designed to study their effect on the corrosion behaviour of steel.
2. Similar corrosion rate for all samples in 3.5% NaCl was obtained from the Tafel extrapolation method owing to the very aggressive chloride environment where the variation of alloying elements in the steel is not enough to influence corrosion process.
3. Atmospheric exposure condition provided good results for the developed steel. The rust resistance comparable to the diameter of the semicircle of Nyquist plots of developed steel G22 was estimated to be at least three times higher than LC and nearly similar to that of SCOR and reference steel SAILCOR.

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