

Corrosion Prevention of Cast Iron Industrial Water Pipes: A Preliminary Comparative Study of Hexamine and Aniline Inhibitors

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Abstract

Using cast iron pipes in various industrial and water systems is experiencing a major problem of corrosion occurrence. Hence the operation and maintenance of these pipes become costly and infeasible. Corrosion inhibitors have a great role in decreasing pipes corrosion rate. In this study, the inhibition effect by applying two inhibitors of hexamethylenetetramine (hexamine) and aniline on cast iron pipes was studied. Experimental measurements of the corrosion behavior of cast iron pipes was thoroughly examined in three aqueous salt solutions of 2% NaCl, 2% Na₂SO₄ and 2% CaCO₃. The corrosion inhibition efficiency of the cast iron pipes by aniline or hexamine in the three aqueous salt aqueous solutions was investigated at constant temperature and for different time intervals. Corrosion rates of the pipes were determined using weight loss technique. It has been found that, for the corrosion of cast iron pipes, a satisfactory inhibition efficiency is observed for a concentration close to 150 ppm hexamine and 150 ppm aniline over the whole aqueous salt solutions tested in the work. The results showed that at the same inhibitor concentration and temperature, aniline exhibits higher inhibition corrosion efficiency on cast iron pipes than the efficiency achieved by hexamine.

Keywords: cast iron pipes; corrosion inhibitors, hexamethylenetetramine; aniline; aqueous salt solution

1. Introduction

Cast iron as an alloy is widely used for water carrying purposes besides mild steel and other metals. Cast iron is also widely used in industrial water piping systems for more than one century. In the past, in industry the pipes were used especially for carrying water were made of cast iron (Mohebbi and Li 2011). The extent and cost of damage caused by corrosion in cast iron water pipes has been rising during recent decades (Mehra and Soni 2002). The use of cast iron in industrial water pipes and potable water distribution systems is essentially suffering from an inevitable corrosion problems

(Atkinson et al. 2002). Actually, this corrosion phenomenon is now considered as the main problem facing cast iron water pipes operation and maintenance in industry, potable water distribution, and wastewater systems (Agatemor and Okolo 2008; Daneshvar-Fatah et al. 2013; Hasan and Sadek 2014; Li et al. 2016; Liang et al. 2013). The corrosion of cast iron pipes is actually varied regarding both material quality and purpose of using (Yang et al. 2012). Corrosion leads to deterioration and failures of those industrial pipes and equipment made of cast iron (Essa 2006; Kuźnicka 2009). The high cost of occurring corrosion in industry and water systems shows the need to improved corrosion measurement and prevention schemes (Reynaud 2010).

Among available solutions of corrosion in engineering materials, inhibitors were found to be of high practical importance, in minimizing metallic waste (Collins et al. 1993). Corrosion protection aims to improve performance of pipes metal (Dwivedi et al. 2017). Methods of Corrosion control are needed to be properly selected according to environment and operational conditions of pipes and equipment (Mannivanan et al. 2012). Corrosion inhibitors are employed as it has been observed the absence of corrosion inhibitors leads always to an exponential increase in corrosion rate of pipe metals (Barmatov et al. 2015). Corrosion inhibitors are commonly single organic components, but mixtures of solvents-compound or compound-surfactant are regularly used (Finšgar and Jackson 2014; Hill and Jones 2003). Various nitrogen or sulfur-containing organic compounds have been used as corrosion inhibitors (Al-Rawajfeh and Al-Shamaileh 2007; Ebenso et al. 2001; Ekpe et al. 1995; Fathima Sabirneeza et al. 2015; Hosseini et al. 2003). The mechanism of corrosion inhibition in surface processes involves adsorption of the inhibitor organic compounds on the metal surface that needed to be protected (Zhu et al. 2015). Inhibition efficiency of organic compounds is usually depends on inhibitor molecular size and the mode of interaction with metal surface (Shirazi et al. 2017).

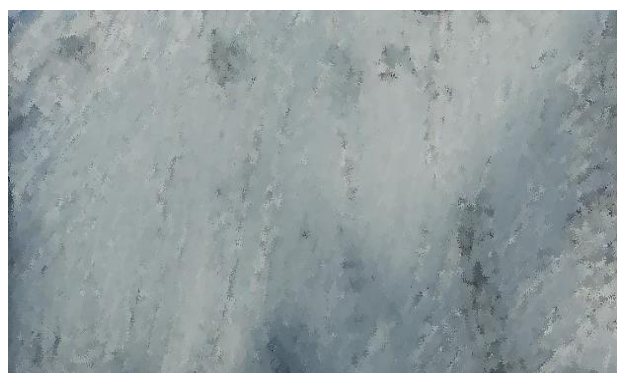
The corrosion of cast iron in acidic and alkaline mediums was studied in several previous works (Osarolube et al. 2008; Simsek et al. 2010). These studies figured out aqueous salt solutions, at high salt concentrations such as 3.0 M, are the most corrosive for cast iron metal. The corrosion behavior was characterized by two factors of salt and oxygen dissolved in aqueous solutions (Shakir et al. 2018). Many previous works have studied hexamine (hexamethylenetetramine) and aniline or their derivatives inhibition properties to protect metals in different acidic and alkaline mediums. The studies were made for diverse metals and alloys such as copper and iron (Benchikh et al. 2009; Essa 2007; Khaled and Hackerman 2004; Vashi and Naik 2010). It has been found that low

molecular mass and high water solubility amines such as hexamine produce higher adsorption and corrosion prevention (Bayol et al. 2007). Aniline and its derivatives are also used as inhibitors as they found to inhibit metal corrosion, especially iron with great extent (Jeyaprabha et al. 2006).

As any obtained information on the rate at which corrosion initiates and progress in cast iron pipes is considered to be important for the attempts to control or reduce the damage caused by corrosion. Moreover, the controlling of deterioration and failures become extremely challenging without a well understanding of the cast iron pipes corrosion. In this work, the aim is to investigate the inhibition effect of aniline and hexamine on cast iron pipes in three different aqueous salt solutions NaCl, Na₂SO₄, and CaCO₃. The corrosion rate of cast iron pipe was experimentally determined using weight loss method with and without inhibitors presence.

2. Materials and Method

2.1. Material Preparation and Weight Loss Measurement Specimens were cut from cast iron water pipes of outer diameter 24 mm, a thickness of 2 mm. The arrangement of the cast iron alloy testing was as coupon specimen of 2x2 cm² and thickness 0.2 cm, a hole was drilled diameter 0.05 cm at the upper edge. The surface of specimens were cleaned, degreased in benzene, washed using 50% acetone, dried, marked and weighed to a constant weight before exposing to the corrosive medium. The specimens were suspended by a glass hook in a beaker filled with test solution, for different duration of immersion 72, 120, 168, 240, and 288 hours in three aqueous salt corrosive mediums. All test solutions were prepared from analytical grade reagents and double – distilled water. The testing aqueous salt solutions are 2% NaCl, 2% Na₂SO₄ and 2% CaCO₃ at room temperature. At the end of each exposure time, the specimens were removed, cleaned, dried and weighed. All specimen metal surfaces, including the edges, were abraded to original ground using grit silicon carbide papers to remove any coated layer to prevent corrosion to pipes like galvanized zinc layer. Figure 1 (a) shows the clean surface of specimen cast iron pipe, as can be seen in Figure 1 (b), the specimen cast iron pipe with a localized corrosion condition is being evident on pipe surface after 288-hour (12 days) immersion in 2% NaCl solution.



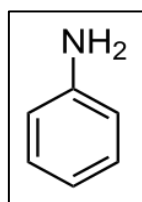
(a)



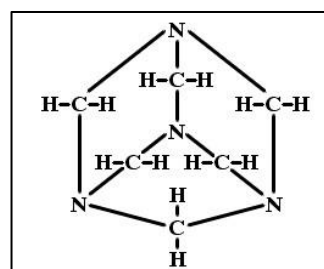
(b)

Figure 1.a. The clean surface of specimen cast iron pipe, Figure 1.b. The specimen cast iron pipe with localized corrosion on the surface after 12 days immersion in 2% NaCl solution.

The chemicals aniline and hexamine were used as corrosion inhibitors for this investigation. The inhibitors concentrations were 150 ppm were prepared in 2% NaCl, 2% Na₂SO₄ and 2% CaCO₃ aqueous salt solutions at 25 ± 2 °C. The molecular structures of the inhibitors used are displayed in Figure 2 (a), and (b).



(a)



(b)

Figure 2.a. the molecular structure of the aniline inhibitor. Figure 2.b. the molecular structure of the hexamethylenetetramine (hexamine) inhibitor.

2.2. Pipe Metal Analysis

The cast iron pipe specimens were analyzed using ICP-OES: Spectro Arcos in the chemical laboratory at University of Garmian. The chemical composition of the cast iron pipes is shown in Table 1. The instrument conditions used were: Spray chamber is Scott spray; Nebulizer: crossflow; RF power/W: 1400; pump speed: 30 RPM; Coolant flow (L/min): 14; Auxiliary flow (L/min): 0.9; nebulizer gas flow (L/min): 0.8; Preflush (s): 40; Measure time (s): 28; replicate measurement: 3; argon gas (purity ≥ 99.99); multi-elements stock solutions containing 1000 mg/L were obtained from Bernd Kraft (Bernd Kraft GmbH, Duisburg, Germany); standard solutions were diluted by several dilution in 0.5% nitric acid as diluent.

Table 1. Chemical composition of cast iron pipes used for water carrying

Component*	Fe	Si	Mn	S	Ni	Cu	Pb	Mo	V	Mg	Cr
Wt. Percentage (%)	95.00	1.00	1.70	0.02	0.06	0.06	0.01	0.03	0.06	0.02	0.14

*The rest is carbon C.

2.3. Inhibition Efficiency and Degree of Surface Coverage Calculations

After the weight loss of cast iron specimens, efficiency was determined as the difference in the weight before and after each exposure time in test aqueous salt solutions for each inhibitor. The values of percentage corrosion inhibition efficiency of aniline and hexamine inhibitors in the three investigated aqueous salt solutions for the various immersion periods was calculated using the following equation that obtained from literature (Abiola et al. 2013; James and Akaranta 2011; Rafiquee et al. 2009). The definition of each symbol in the following equation is presented in the nomenclature at the end of this paper.

$$IE\% = \left(1 - \frac{W_i}{W_n}\right) * 100\% \quad (1)$$

The degree of surface coverage, θ was determined by the following equation (Daoud et al. 2015; Sirajunnisa et al. 2014);

$$\theta = \left(1 - \frac{W_i}{W_n}\right) \quad (2)$$

2.4. Corrosion Rate Calculations

The corrosion rate of cast iron in different aqueous salt solution mediums was determined for different immersion period from weight loss using the equation below. The same corrosion rate (CR) equation was used for various metals and solutions (Anand and Balasubramanian 2011; Singh and Quraishi 2015):

$$C_R = \frac{87.6 W}{AtD} \quad (3)$$

Where W (in mg) is the weight loss and calculated as follows:

$$W = W_0 - W_t \quad (4)$$

3. Results and Discussion

3.1. Morphology of Corroded Metal Surface

The corroded metal surface has been changed in appearance and its color was turned into brown for all the specimens of cast iron pipes. As shown in Figure 1 typical changes in the corrosion products on a specimen after 12 days exposure in 2% NaCl. In general, the look of the corroded surface of cast iron specimens was the same for all the investigated aqueous salt solutions, but the thickness of corrosion product varies with exposure time. The localized corrosion occurred on specimen surface is the main form of corrosion of water used cast iron pipes.

3.2. Weight Loss and Corrosion Rates

Results obtained from weight loss and corrosion rate of cast iron pipe specimens in a 2% NaCl, 2% Na₂SO₄, and 2% CaCO₃ solutions at 25±2°C are showed in Tables 2 and 3. From which it can be observed that the weight loss for specimens in the three solutions increases with time, in consequence, the corrosion rate is also increases with time. The corrosion rate of cast iron specimens in the test solutions was calculated from the decrease in weight loss by applying equations 3 and 4.

Table 2. Weight loss (mg / cm²) of the cast iron specimens in 3 aqueous salt solutions of 2% NaCl, 2% ppm Na₂SO₄ and 2% ppm CaCO₃ at 25±2°C and for different time intervals.

Time (hr.)	24	72	120	168	240	288
NaCl	0.138	0.422	0.774	1.181	1.689	1.882
Na ₂ SO ₄	0.059	0.248	0.493	0.731	0.974	1.134
CaCO ₃	0.057	0.191	0.322	0.484	0.668	0.748

Table 3. Corrosion rates (mmpy) of the cast iron specimens in three aqueous salt solutions of 2% NaCl, 2% ppm Na₂SO₄ and 2% ppm CaCO₃ at 25±2°C and for different time intervals.

Time (hr.)	24	72	120	168	240	288
NaCl	0.0690	0.0704	0.0774	0.0844	0.0845	0.0784
Na ₂ SO ₄	0.0295	0.0414	0.0493	0.0522	0.0487	0.0473
CaCO ₃	0.0284	0.0318	0.0322	0.0346	0.0334	0.0312

For the specimens in 2% NaCl solution, the corrosion rate seems to follow a specific trend and it appears to increase with time. But this increase is more drastic and then tends to be less at longer exposure time. For the specimens in 2% Na₂SO₄ solution, the corrosion rate seems to establish a different trend as it decreases at the higher exposure time. For the specimens in 2% CaCO₃ solution, the average corrosion rate is close at exposure times longer than 120 hr. Based on the analysis of the corrosion rate results obtained from the three aqueous salt solutions it can be understand that the localized

corrosion behavior of cast iron water pipes is the primary form of corrosion degradation. The intensity of the localized corrosion depends on the extent of time exposure. From Tables 2 and 3, where the weight loss and corrosion rate values were listed, it can be observed from corrosion rate in mmpy of cast iron specimens in the three tested aqueous salt solutions are in the order of $\text{NaCl} > \text{Na}_2\text{SO}_4 > \text{CaCO}_3$ during time of exposure of 288 hours.

3.3. Effect of Corrosion Inhibitors

The weight loss measurements were carried out of cast iron specimens with aniline and hexamine corrosion inhibitors concentrations of 150 ppm separately. The exposure time was ranging from 24 to 288 hours to study the effect of inhibitor presence and immersion time on the corrosion rate of cast iron water pipes at $25 \pm 2^\circ\text{C}$ as seen in Table 4 and 5. It was found that with use of 150 ppm concentration of aniline and hexamine inhibitors causes decreasing of weight loss in all the studied aqueous salt solutions as displayed in Figure 3 to 5.

Figure 3 shows the results of specimen weight loss produced from corrosion of cast iron pipes for different exposure time in three conditions of 2% NaCl aqueous salt solution. In one these conditions 150 ppm of hexamine inhibitor was added and in another, a 150 ppm aniline was added to the solution.

As illustrated in Figure 3, the amount of weight loss was decreased considerably when 150 ppm hexamine was added to NaCl solution. The weight loss of cast iron specimens was more significantly decreased when 150 ppm aniline was added to the solution. The same effect was noticed for the other two aqueous solutions of 2% Na_2SO_4 and 2% CaCO_3 as presented in Figures 4 and 5 for the exposure time ranges from 24 hr. to 288 hr.

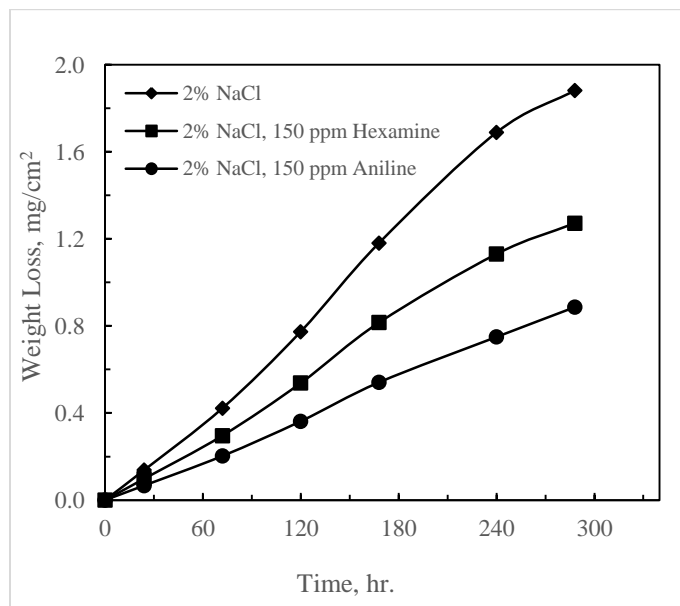


Figure 3. Variation of specific weight loss with time of cast iron specimens in 150 ppm Aniline and 150 ppm hexamine inhibitors added to 2% NaCl solution at $25 \pm 2^\circ\text{C}$.

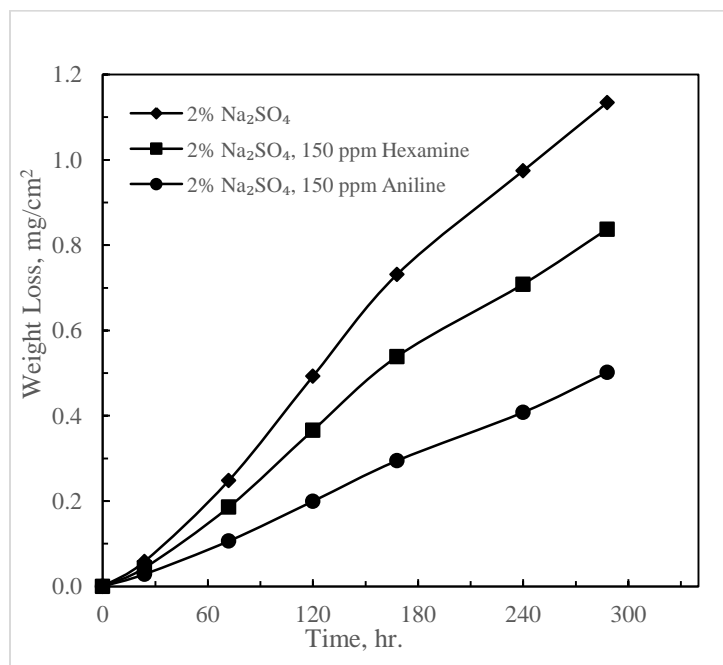


Figure 4. Variation of specific weight loss with time of cast iron specimens in 150 ppm Aniline and 150 ppm hexamine inhibitors added to 2% Na₂SO₄ solution at $25 \pm 2^\circ\text{C}$.

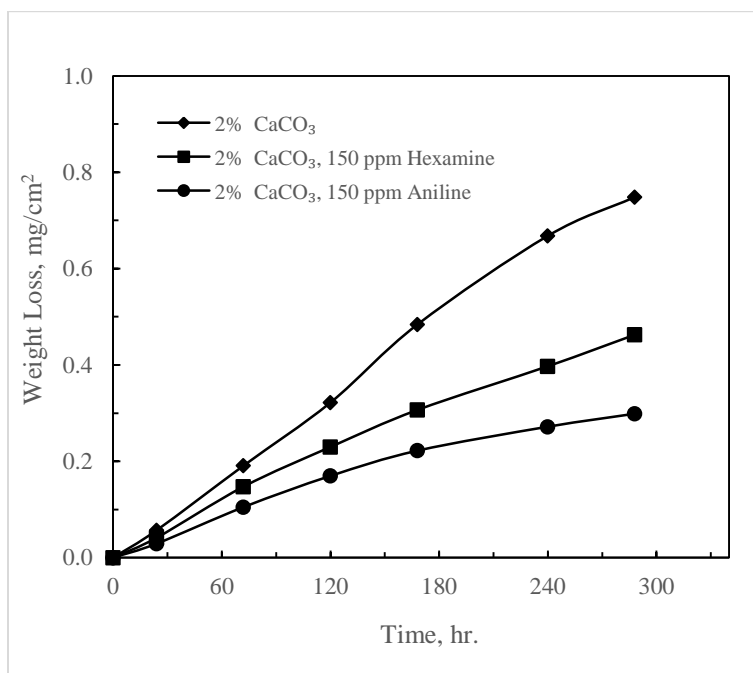


Figure 5. Variation of specific weight loss with time of cast iron specimens in 150 ppm Aniline and 150 ppm hexamine inhibitors added to 2% CaCO₃ solution at 25±2°C.

From Table 4 it was found that with increase in exposure time from 24 to 288 hours, the weight loss decreased and hence the inhibition efficiency increased from 28.95% to 32.41% when 150 ppm hexamine was added to the solution of 2% NaCl (surface coverage increased from 0.289 to 0.324). The increase ranges of efficiency of 2% Na₂SO₄ and 2% CaCO₃ were (from 25.46% to 26.16%) and (from 27.37% to 35.38%).

In terms of metal protection, these results indicate that adding 150 ppm of hexamine is the satisfactory concentration to develop acceptable corrosion prevention for cast iron pipe specimens in the investigated salt solutions. The behavior of hexamine inhibitor most probably results from adsorption on metal surface is suitable with this concentration of hexamine and therefore the inhibition efficiency was reasonable.

Table 4 shows the results of 150 ppm aniline added to the tested aqueous salt solutions. It can be observed that the corrosion rate reduced during with time interval from 24 to 288 hours for 2% NaCl, 2% Na₂SO₄, and 2% CaCO₃. The reduction of corrosion rate increases higher than that achieved by 150 ppm hexamine. The reason of this difference is most possibly due to the adsorption behavior of anions the electrolyte of aniline at the electrode surface (Luo et al. 1998).

Table 4. Corrosion parameters, obtained from weight loss measurements for cast iron specimens in three aqueous salt solutions of 2% NaCl, 2% Na₂SO₄ and 2% CaCO₃ at 25± 2°C, containing 150 ppm hexamine inhibitor for different time intervals.

Exposure time (hr.)	Corrosion rate (mmpy)	Inhibition efficiency (%)	Surface coverage (θ)
2%NaCl			
24	0.0490	28.95	0.289
72	0.0494	29.86	0.299
120	0.0538	30.52	0.305
168	0.0583	30.96	0.310
240	0.0565	33.05	0.331
288	0.0530	32.41	0.324
2% Na ₂ SO ₄			
24	0.0220	25.46	0.255
72	0.0310	25.13	0.251
120	0.0366	25.70	0.257
168	0.0385	26.31	0.263
240	0.0354	27.27	0.273
288	0.0349	26.16	0.262
2% CaCO ₃			
24	0.0206	27.37	0.274
72	0.0246	22.83	0.228
120	0.0230	28.66	0.287
168	0.0219	36.65	0.366
240	0.0209	37.53	0.375
288	0.0202	35.38	0.354

From the results presented in Table 4, it was found with the weight loss increases with time for all three solutions and the corrosion rate remains at certain levels with time for NaCl and Na₂SO₄ solutions. However, for 2% CaCO₃ solution, the corrosion rate drops relatively after 168 hours. This declining is perhaps because of the stability of ferric oxide film that formed after corrosion occurrence. The phenomena is known for corrosion inhibitor behavior of anions in aqueous solutions (Xu et al. 2017). In general, it is noticed that the corrosion rate is highly reduced in the solutions contain hexamine or aniline inhibitor. The reduction occurs due to the inhibitor protection for the metals by preventing the direct contact between metals surface and corrosive anions. In the same time, the reduction is also made by decreasing plenty of cations on the metal surface. At 25 °C ± 2 and 240 hours with presence 150 ppm hexamine and 150 ppm aniline, the

corrosion rate of 2% NaCl solution was lessened to 0.0565 and 0.0375 mmpy respectively. While for Na₂SO₄ and CaCO₃ solutions, same thing happened at varying rates of the corrosion. The two anions are reported to have a significant influence on the corrosion characteristics of cast iron are chloride and sulfate ions (Ekpe et al. 2001).

Table 5. Corrosion parameters obtained from weight loss measurements for cast iron specimens in three aqueous salt solutions of 2% NaCl, 2% Na₂SO₄ and 2% CaCO₃ at 25±2°C, containing 150 ppm aniline inhibitor for different time intervals.

Exposure time (hr.)	Corrosion rate (mmpy)	Inhibition efficiency (%)	Surface coverage (θ)
2% NaCl			
24	0.0335	51.50	0.515
72	0.0338	52.00	0.520
120	0.0362	53.20	0.532
168	0.0386	54.20	0.542
240	0.0375	55.62	0.556
288	0.0369	52.90	0.529
2% Na ₂ SO ₄			
24	0.0144	51.08	0.511
72	0.0177	57.20	0.572
120	0.0200	59.50	0.595
168	0.0211	59.70	0.597
240	0.0204	58.12	0.581
288	0.0209	55.77	0.558
2% CaCO ₃			
24	0.0145	48.79	0.488
72	0.0175	45.06	0.451
120	0.0170	47.30	0.473
168	0.0159	54.12	0.541
240	0.0136	59.31	0.593
288	0.0124	60.08	0.601

The exposure time effect on cast iron corrosion rate from 24 to 288 hours was also explored in this work. For both hexamine and aniline inhibitor in 2% NaCl solution, the inhibition efficiency enhanced (from 28.95 % to 32.41 %), and (from 51.50 % to 52.90 %) respectively.

The increase in inhibition efficiency at longer immersion time is due to anion kinetics in aqueous solutions of strengthening of adsorption that mentioned above. The immersion time with corrosion inhibitors aniline and hexamine scores highest inhibition efficiency

of cast iron specimens in 2% NaCl, 2% Na₂SO₄ and 2% CaCO₃ at 288 hours as given in Tables 3 and 4.

As a result, the inhibition increased as more inhibitor molecules are adsorbed on the metal surface reduces the surface area available for the attack of the aggressive ions from the salt solution. Inhibition efficiency showed remarkable improvement with addition of 150 ppm to 2% CaCO₃ probably for the same reason mentioned above.

Conclusions

The inhibition of corrosion of cast iron alloy by an addition of 150 ppm hexamine and 150 ppm aniline was performed efficiently in salts solutions of 2% NaCl, 2% Na₂SO₄ and 2% CaCO₃ at 25±2 °C.

The aniline showed more effective inhibition efficiency than the hexamine. In 2% NaCl solution the corrosion rate of cast iron pipes was higher than 2% Na₂SO₄ and 2% CaCO₃ solutions.

Generally, in aqueous salt solutions, the corrosion rate of cast iron alloy appears to be a function of dissolved salt type, inhibitor type and immersion time.

The work described here lead us to expect the using aniline as a corrosion inhibitor rather than hexamine in prevention the cast iron corrosion in industry, where cast iron pipes still implemented to carry aqueous salt solutions.

Nomenclature

- A the area of the specimen (cm²),
C_R the corrosion rate (CR) of cast iron (mmpy),
t the exposure time (h),
D the density of cast iron (g/cm³)
IE% the inhibition efficiency (%)
θ the surface coverage (-)
W weight loss (mg),
W_i the weight loss of cast iron in the corrodent – inhibitor system (mg),
W_n the weight loss of cast iron in the corrodent (blank) (mg),
W₀ the weight loss of cast iron in the corrodent (blank) at exposure time 0 (mg),
W_t the weight loss of cast iron in the corrodent (blank) at the end of exposure time (mg),

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