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Corrosion Inhibition of Aluminium by Schiff Base in HNO_3 with and Without Additive (KNO_3)

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Abstract: Weight loss and thermometric methods have been used to study the corrosion inhibition of aluminium in HNO_3 by Schiff base *N*-(4-*N,N*-dimethylaminobenzal)-*p*-anisidine with and without additive (KNO_3). The present study reveals that aluminium in HNO_3 has been more efficiently inhibited by Schiff base in the presence of additive (KNO_3) than Schiff base alone, this enhanced inhibition efficiency in the presence of KNO_3 are due to the synergistic effect. Inhibition efficiency was found maximum (85.69%) for aluminium in 2N HNO_3 by *N*-(4-*N,N*-dimethylaminobenzal)-*p*-anisidine at an inhibitor concentration of 0.7% in presence of additive (KNO_3). Inhibitor adsorbs on the metallic surface and forms a monolayer. Results obtained in both the methods show a good agreement and indicate the dependence of inhibition efficiencies on the concentration of Schiff base, additive (KNO_3) and also on the concentration of HNO_3 solution. The results show the increasing trends of inhibition efficiency with the increasing concentration of inhibitor, additive and also on the increasing concentration of HNO_3 .

Keywords: Weight loss method, Thermometric method, Inhibition efficiency, Corrosion rate, Surface coverage, Synergistic effect

I. INTRODUCTION

The process in which a metal is decayed by a chemical or electrochemical reaction is called corrosion. When metals come into contact with gaseous and liquid environment, the destruction or deterioration of the metals starts at the surface. This type of metallic destruction may be either due to direct chemical attack by the environment [1] or by electrochemical attack.

Corrosion is a natural process whereby metals and alloys return to their unrefined naturally occurring forms as ores and minerals. When ore is refined, this natural and most stable ore is undergoes reverse [2] process to produce the pure metal which is less stable under natural conditions than ore.

Corrosion is almost inevitable in our life. It causes huge losses to buildings, automobiles, industries etc. Aluminum alloys recommended for building purpose are widely used as structural material for various internal outfits in various industries and in highly polluted places, due to its inertness.

Aluminium and its alloys are useful in many engineering applications because of their combination of lightness with strength, their thermal and electrical conductivities, heat and light reflectivity and their hygienic and non toxic qualities. Besides this, aluminium is said to possess an excellent corrosion resistance. This is due to the presence of compact, adherent protective film of aluminium oxide, which isolates aluminium from its environment.

Generally, the organic compounds containing hetero atoms such as N, O, S, Se etc. are found to behave as very effective corrosion inhibitors [3]-[8]. The inhibition efficiency of these compounds depends upon electron density present around the hetero atoms. Inhibition efficiency also depends upon the number of active centers on the surface, charge density, molecular size and mode of adsorption of inhibitor molecules with surface of metal. Corrosion of aluminium and its alloys in different acid media has been extensively studied [9]-[13].

The present investigation deals with the inhibition efficiencies of *N*-(4-*N,N*-dimethylaminobenzal)-*p*-anisidine, with and without additive (KNO_3) in different concentrations of HNO_3 on corrosion of aluminium.

II. EXPERIMENTAL

Rectangular specimens of aluminium of dimension 2.0 cm × 2.0 cm × 0.03 cm containing a small hole of about 2 mm diameter near the upper edge were taken. The chemical composition of the specimen was 98.5% Al, 0.2% Fe, 0.2% Cu, 0.08% Zn and 0.03% Ti. Specimens were cut from a sheet and cleaned by buffing to produce a spotless finish and then degreased. Solutions of HNO_3 and KNO_3 were prepared using double distilled water. Schiff base were synthesised by conventional methods.

Each specimen of aluminium was suspended on a V-shaped glass hook and immersed in a glass beaker containing 50 cc of the test solution at room temperature. The time of exposure of specimen in solution was varied for individual cases. After the exposure, specimens were taken out and cleaned with a saturated solution of ammonium acetate and then dried in hot air. The loss in weight (ΔW) was measured by digital balance having accuracy upto (0.0001g)

The percentage inhibition efficiencies of inhibitors were calculated as [14]:

$$\eta\% = \frac{(\Delta W_u - \Delta W_i)}{\Delta W_u} \times 100$$

Where ΔW_u and ΔW_i are the weight loss of the metal in uninhibited and in inhibited solution, respectively.

The corrosion rate in mm/yr (milli meter per year) was expressed as [15]:

$$\text{Corrosion rate (mm/yr)} = \frac{\Delta W \times 87.6}{A \times T \times d}$$

Where ΔW is the weight loss of specimen in mg, A is the area of exposure of specimen in square cm, T is the time of exposure in hours and d is the density of specimen in g/cm^3 .

The degree of surface coverage (θ) was calculated as [16]:

$$\theta = \frac{(\Delta W_u - \Delta W_i)}{\Delta W_u}$$

Where ΔW_u and ΔW_i are the weight loss of the metal in uninhibited and in inhibited solution, respectively.

Inhibition efficiencies were also calculated using thermometric method. This involves the immersion of a single specimen in an insulating reaction chamber having 50 mL of solution at an initial room temperature. Temperature changes were measured at regular intervals using a thermometer with accuracy of 0.01°C . Initially the increase in temperature was slow, then rapid, attained a maximum value and then decline. The maximum temperature was noted

Percentage inhibition efficiency was calculated as [17]:

$$\eta (\%) = \frac{RN_f - RN_i}{RN_f} \times 100$$

Where RN_f = Reaction Number in the free solution.

RN_i = Reaction Number in the inhibited solution.

Reaction Number, RN (Kmin^{-1}) is given as [18]:

$$RN = \frac{(T_m - T_i)}{t}$$

Where T_m = Maximum temperature of solution.

T_i = Initial temperature of solution.

t= time required (in minutes) to attain maximum temperature.

III.RESULTS AND DISCUSSION

A. Weight loss method

Weight loss (ΔW), percentage inhibition efficiency ($\eta\%$), corrosion rate (mm/yr) and surface coverage (θ) for different concentrations of HNO_3 and those of inhibitors are given in Table-1, Table-2, Table-3 and Table-4. These tables show that inhibition efficiency of inhibitor increases with increasing concentration of inhibitor. Inhibition efficiency also increases with increasing concentration of acid for a given concentration of inhibitor and inhibitor show maximum inhibition efficiency at the highest concentration of acid used i.e. 2.0N HNO_3 . Corrosion rate increases with the increase in concentration of acid without inhibitor and decreases with the increase in concentration of inhibitor in an acid. The value of surface coverage increases with increasing concentration of inhibitor. Surface coverage also increases with increasing concentration of acid and the inhibitor show maximum surface coverage at the highest concentration of acid used i.e. 2.0N HNO_3 . The maximum inhibition efficiency (i.e. 78.32%) was obtained at an inhibitor concentration of 0.7% in 2.0N HNO_3 . The maximum corrosion rate (i.e. 3.09mm/yr) was obtained for uninhibited solution in 2.0N HNO_3 . The maximum surface coverage (i.e. 0.78) was obtained at an inhibitor concentration of 0.7% in 2.0N HNO_3 .

The variation of percentage inhibition efficiency with inhibitor concentration is presented graphically in fig.1 for 0.1N HNO₃, fig.2 for 0.5N HNO₃, fig.3 for 1.0N HNO₃ and fig.4 for 2.0N HNO₃. Figures show a linear curve of percentage inhibition efficiency with inhibitor concentration, which indicates that the inhibition efficiency increases linearly with increasing inhibitor concentrations.

Weight loss, percentage inhibition efficiencies, corrosion rate and surface coverage for different concentrations of acid and those of inhibitor in presence of different concentrations of additive (KNO₃) are given in Table-5, Table-6, Table-7 and Table-8. Tables show the same trend as observed in HNO₃ alone but with higher values of inhibition efficiencies. Inhibition efficiency also increases with increasing concentration of acid as well as that of additive (KNO₃) and the inhibitor show maximum inhibition efficiency at the highest concentration of acid (i.e. 2.0N HNO₃) in presence of highest concentration of additive (KNO₃) i.e. 2.0N KNO₃. Surface coverage also increases with increasing concentration of acid as well as that of additive (KNO₃) and the inhibitor show maximum surface coverage at the highest concentration of acid used (i.e. 2.0N HNO₃) in presence of highest concentration of additive (KNO₃) i.e. 2.0N KNO₃. The maximum inhibition efficiency (i.e. 85.69%) was obtained at an inhibitor concentration of 0.7% in 2.0N HNO₃ in presence of additive 2.0N KNO₃. The maximum surface coverage (i.e. 0.86) was obtained at an inhibitor concentration of 0.7% in 2.0N HNO₃ in presence of additive (KNO₃). The variation of percentage inhibition efficiency with concentration of inhibitor in presence of KNO₃ is presented graphically in fig.5 for 0.1N HNO₃, fig.6 for 0.5N HNO₃, fig.7 for 1.0N HNO₃ and fig.8 for 2.0N HNO₃. Figures show a linear curve of percentage inhibition efficiency with concentration of inhibitor in presence of KNO₃, which indicates that the inhibition efficiency increases linearly with increasing inhibitor concentrations in presence of KNO₃.

The present study revealed that aluminium in HNO₃ has been more efficiently inhibited by Schiff's bases in presence of KNO₃ than Schiff's bases alone. This is due to the synergistic effect, the effect of two chemicals on a metal surface to be greater than the effect of each chemical individually or the sum of their individual effects.

B. Thermometric Method

Inhibition efficiencies were also determined using thermometric method. Temperature changes for test solutions in 1.0N, 2.0N and 3.0N HNO₃ were recorded both in presence and in absence of the additive (KNO₃). However, no significant temperature changes were recorded in 0.1N and 0.5N concentrations of the acid. Results summarised in Table-9 for HNO₃ and in Table-10 for HNO₃ in presence of additive (KNO₃) show a good agreement with the results obtained by weight loss method. The maximum inhibition efficiency (i.e. 79.79%) in table-9 was obtained with the highest concentration (i.e. 0.7%) of inhibitor and with highest concentration of HNO₃ (i.e. 3.0N) and in table-10 was obtained (i.e. 84.57%) with the highest concentration (i.e. 0.7%) of inhibitor and with highest concentration of HNO₃ (i.e. 3.0N) in presence of additive (KNO₃). The variation of reaction number (RN) with inhibitor concentration is depicted graphically in Fig.9 for 3.0N HNO₃ and in Fig.10 for 3.0N HNO₃ in presence of additive 3.0N KNO₃. Figures show a linear deviation of reaction number with the concentration of inhibitor which indicates that the reaction number decreases with increasing concentration of inhibitor. Generally, organic molecules containing hetero atoms such as N, O, S adsorb on the metal surface and inhibit the surface corrosion. In the case of Schiff base nitrogen atom is responsible for adsorption. Nitrogen atom of Schiff base which are adsorb on the metal surface form a monolayer, thus causes a decrease in corrosion rate. In the present investigation N-(4-N,N-dimethylaminobenzal)-p-anisidine shows maximum inhibition efficiency, this may be due to its molecular structure. The -OCH₃ group present in this Schiff base exerts a positive mesomeric effect (+M>I) which increases the electron density at the nitrogen atom. This increased electron density at nitrogen atom offers better adsorptivity of the Schiff Base on the corroding sites of the metal. It has also been observed that the inhibition efficiency of inhibitors is higher in higher concentration of HNO₃. This may be because of the fact that the inhibitor ionises more readily under more acid strength and is adsorbed more easily on the metal surface. Therefore, they act as better inhibitors at higher concentrations of acids. A comparison of inhibition efficiency of newly synthesised Schiff base with and without additives has been studied. The present study has revealed that aluminium in nitric acid has been more efficiently inhibited by Schiff base with additive (KNO₃). This is due to the synergistic effect. The effect of two chemicals on metal surface to be greater than the effect of each chemical individually or the sum of their individual effects. The enhanced inhibition efficiency in the presence of nitrate ion is only due to the synergism of nitrate ion. Adsorption plays an important role in the inhibition of metallic corrosion by organic inhibitors. The efficiencies of inhibitors expressed as the relative reduction in corrosion rate can be qualitatively related to the amount of adsorbed inhibitors on the metal surface. It is assumed that the corrosion reactions are prevented from occurring over the active sites of the metal surface covered by adsorbed inhibitors species, whereas, the corrosion reaction occurs normally on the surface at inhibitors free area. The inhibition efficiency is thus, directly proportional to the fraction of the surface covered with adsorbed inhibitors.

IV. CONCLUSION

- A. Weight loss method has shown that the inhibition efficiency of Schiff base increases with increasing concentration of inhibitor and with increasing concentration of acid.
- B. Weight loss method was carried out in presence of additive it has also shown that inhibition efficiency of Schiff base also increases with increasing concentration of KNO_3 as additive.
- C. The compound under investigation which show the highest inhibition efficiencies (i.e. 78.32% in 2.0N HNO_3) at an inhibitor concentration of 0.7% in absence of 2.0N KNO_3 and inhibitor shows (i.e. 85.69% in 2.0N HNO_3) at an inhibitor concentration of 0.7% in presence of 2.0N KNO_3 . Such efficiencies are higher than the efficiencies of Schiff base alone.
- D. The present study shows that aluminium in HNO_3 has been more efficiently inhibited by Schiff base in the presence of additive (KNO_3) than Schiff base alone due to the synergistic effect between Schiff base and KNO_3 .
- E. Results of inhibition efficiencies observed from weight loss and thermometric methods have shown fairly good agreement.
- F. Thermometric method has not applicable on the lower concentration of acids 0.1N and 0.5N acid solutions.
- G. The inhibitor functions by forming a barrier between metal and corrosive medium through chemisorbtion.

Table – 1

Weight loss (ΔW) and percentage inhibition efficiency ($\eta\%$) for Aluminium in HNO_3 solution with inhibitor.

Inhibitor concentration	0.1N HNO_3 (192hrs.)			
	ΔW (mg)	I.E. ($\eta\%$)	Corrosion rate (mm/yr)	Surface Coverage (θ)
Uninhibited	23.1	-	0.53	-
Schiff Base				
0.1%	13.3	42.42	0.30	0.42
0.3%	12.7	45.02	0.29	0.45
0.5%	11.3	51.08	0.26	0.51
0.7%	9.7	58.01	0.22	0.58

Table – 2 Weight loss (ΔW) and percentage inhibition efficiency ($\eta\%$) for Aluminium in HNO_3 solution with inhibitor.

Inhibitor concentration	0.5N HNO_3 (144hrs.)			
	ΔW (mg)	I.E. ($\eta\%$)	Corrosion rate (mm/yr)	Surface Coverage (θ)
Uninhibited	37.2	-	1.13	-
Schiff Base				
0.1%	18.7	49.73	0.57	0.50
0.3%	17.3	53.49	0.53	0.53
0.5%	15.7	57.80	0.48	0.58
0.7%	12.7	65.86	0.39	0.66

Table – 3

Weight loss (ΔW) and percentage inhibition efficiency ($\eta\%$) for Aluminium in HNO_3 solution with inhibitor.

Inhibitor concentration	1.0 N HNO_3 (96 hrs.)			
	ΔW (mg)	I.E. ($\eta\%$)	Corrosion rate (mm/yr)	Surface Coverage (θ)
Uninhibited	44.9	-	2.05	-
Schiff Base				
0.1%	20.1	55.23	0.92	0.55
0.3%	18.7	58.35	0.85	0.58
0.5%	17.0	62.14	0.77	0.62
0.7%	14.3	68.15	0.65	0.68

Table – 4

Weight loss (ΔW) and percentage inhibition efficiency ($\eta\%$) for Aluminium in HNO_3 solution with inhibitor.

Inhibitor concentration	2.0 N HNO_3 (96 hrs.)			
	ΔW (mg)	I.E. ($\eta\%$)	Corrosion rate (mm/yr)	Surface Coverage (θ)
Uninhibited	67.8	-	3.09	-
Schiff Base				
0.1%	25.3	62.68	1.15	0.63
0.3%	22.9	66.22	1.04	0.66
0.5%	19.3	71.53	0.88	0.72
0.7%	14.7	78.32	0.67	0.78

Table – 5 Weight loss (ΔW) and percentage inhibition efficiency ($\eta\%$) for Aluminium in HNO_3 solution with inhibitor in presence of additive (KNO_3).

Inhibitor concentration	0.1N HNO_3 + 0.1N KNO_3 (192 hrs.)			
	ΔW (mg)	I.E. ($\eta\%$)	Corrosion rate (mm/yr)	Surface Coverage (θ)
Uninhibited	23.1	-	0.53	-
Schiff Base				
0.1%	12.3	46.75	0.28	0.47
0.3%	11.1	51.95	0.25	0.52
0.5%	9.9	57.14	0.22	0.57
0.7%	7.9	65.80	0.18	0.66

Table – 6

Weight loss (ΔW) and percentage inhibition efficiency ($\eta\%$) for Aluminium in HNO_3 solution with inhibitor in presence of additive (KNO_3).

Inhibitor concentration	0.5N HNO_3 + 0.5N KNO_3 (144 hrs.)			
	ΔW (mg)	I.E. ($\eta\%$)	Corrosion rate (mm/yr)	Surface Coverage (θ)
Uninhibited	37.2	-	1.33	-
Schiff Base				
0.1%	15.7	57.80	0.48	0.58
0.3%	14.2	61.83	0.43	0.62
0.5%	12.3	66.94	0.37	0.67
0.7%	10.3	72.31	0.31	0.72

Table – 7

Weight loss (ΔW) and percentage inhibition efficiency ($\eta\%$) for Aluminium in HNO_3 solution with inhibitor in presence of additive (KNO_3).

concentration	1.0 N HNO_3 + 1.0 N KNO_3 (96 hrs.)			
	ΔW (mg)	I.E. ($\eta\%$)	Corrosion rate (mm/yr)	Surface Coverage (θ)
Uninhibited	44.9	-	2.05	-
Schiff Base				
0.1%	17.7	60.58	1.24	0.60
0.3%	16.1	64.14	1.31	0.64
0.5%	13.3	70.38	1.44	0.70
0.7%	10.3	77.06	1.58	0.77

Table – 8

Weight loss (ΔW) and percentage inhibition efficiency (η) for Aluminium in HNO_3 solution with inhibitor in presence of additive (KNO_3).

concentration	2.0 N HNO_3 + 2.0 N KNO_3 (96 hrs.)			
	ΔW (mg)	I.E. (η %)	Corrosion rate (mm/yr)	Surface Coverage (θ)
Uninhibited	67.8	-	3.09	-
Schiff Base				
0.1%	22.3	67.11	1.02	0.67
0.3%	19.3	71.53	0.88	0.72
0.5%	15.0	77.88	0.68	0.78
0.7%	9.7	85.69	0.44	0.86

Table-9

Thermometric data for percentage inhibition efficiency \square (%) for Aluminium in 1.0N, 2.0N and 3.0N HNO_3 with inhibitor. Initial temperature – $303 \pm 0.1K$

Inhibitor Concentration C (%)	1.0N HNO_3		2.0N HNO_3		3.0N HNO_3	
	RN ($Kmin^{-1}$)	\square (%)	RN ($Kmin^{-1}$)	\square (%)	RN ($Kmin^{-1}$)	\square (%)
Uninhibited	0.0091	-	0.0133	-	0.0188	-
Schiff Base						
0.1	0.0042	53.85	0.0057	57.14	0.0058	69.15
0.3	0.0039	57.14	0.0050	62.41	0.0054	71.28
0.5	0.0036	60.44	0.0043	67.67	0.0046	75.53
0.7	0.0030	67.03	0.0037	72.18	0.0038	79.79

Table-10 Thermometric data for percentage inhibition efficiency \square (%) for Aluminium in 1.0N, 2.0N and 3.0N HNO_3 with inhibitor in presence of additive (KNO_3). Initial temperature – $303 \pm 0.1K$

Inhibitor Concentration C (%)	1.0N HNO_3 + 1.0N KNO_3		2.0N HNO_3 + 2.0N KNO_3		3.0N HNO_3 + 3.0N KNO_3	
	RN ($Kmin^{-1}$)	\square (%)	RN ($Kmin^{-1}$)	\square (%)	RN ($Kmin^{-1}$)	\square (%)
Uninhibited	0.0091	-	0.0133	-	0.0188	-
Schiff Base						
0.1	0.0039	57.14	0.0050	62.41	0.0050	73.40
0.3	0.0036	60.44	0.0047	64.66	0.0046	75.53
0.5	0.0030	67.03	0.0037	72.18	0.0038	79.79
0.7	0.0024	73.63	0.0030	77.44	0.0029	84.57

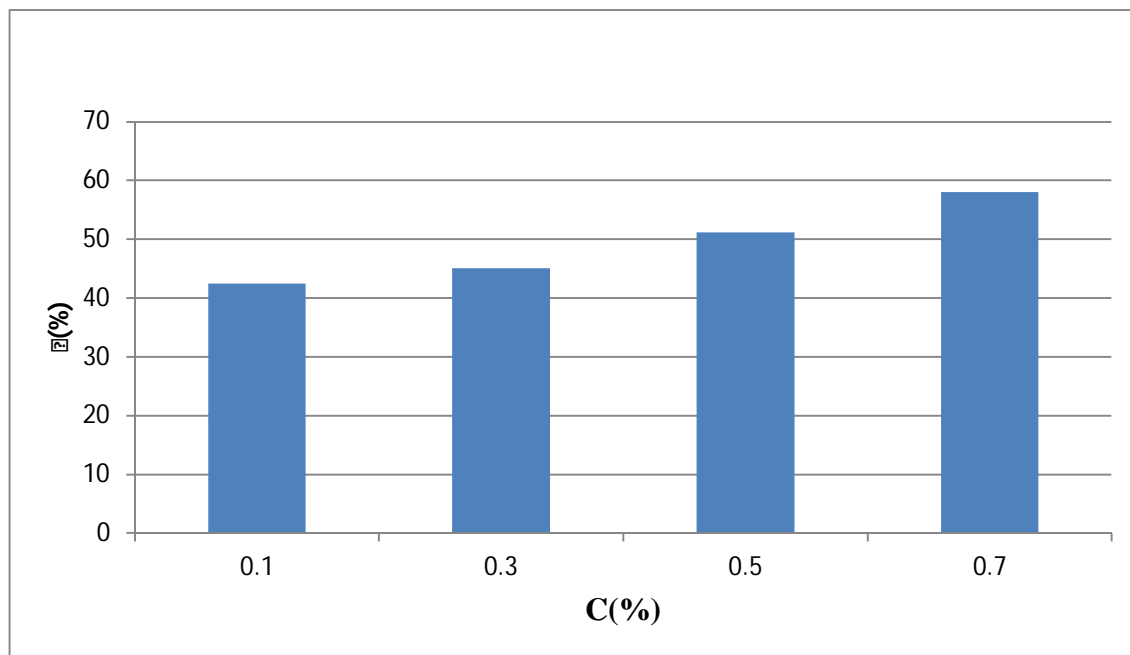


Fig. -1 : - Variation of percentage inhibition efficiency η (%) with inhibitor concentration C (%) for Aluminium in 0.1N HNO_3 .

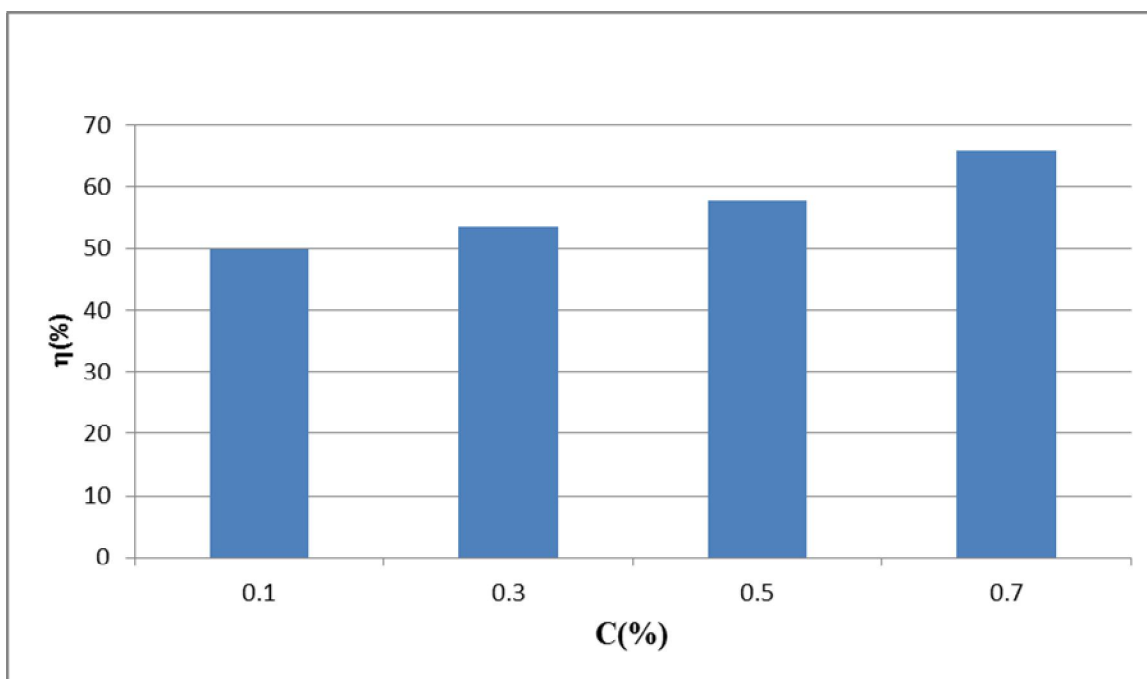


Fig. -2 : - Variation of percentage inhibition efficiency η (%) with inhibitor concentration C (%) for Aluminium in 0.5 N HNO_3 .

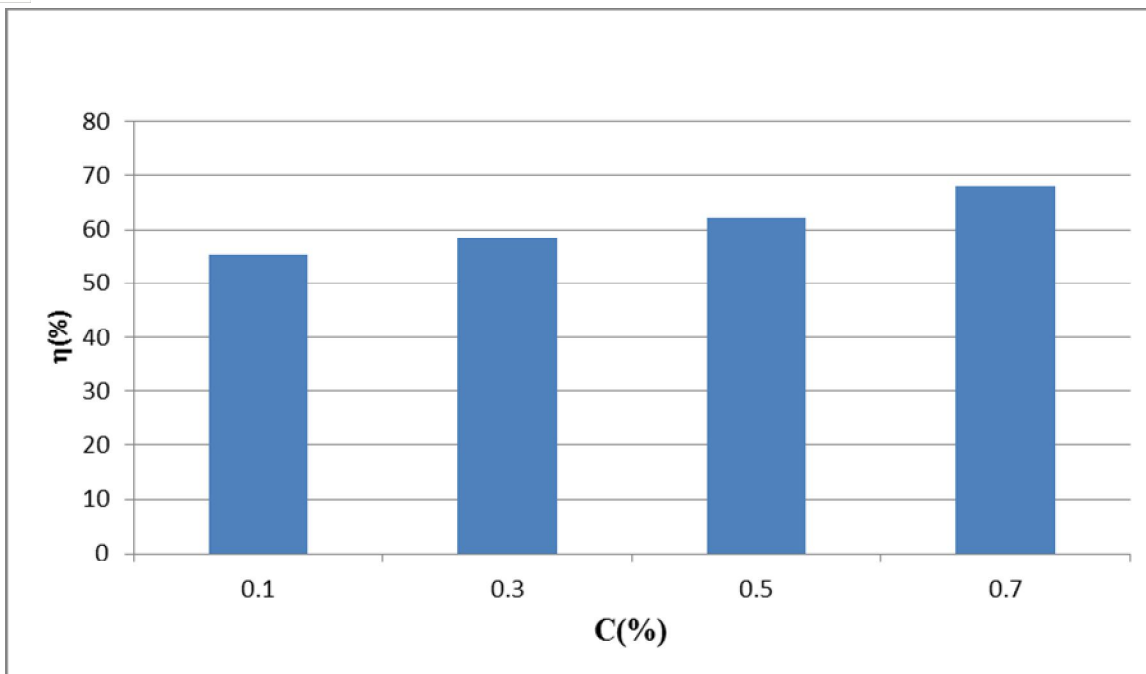


Fig. -3 : - Variation of percentage inhibition efficiency η (%) with inhibitor concentration C (%) for Aluminium in 1.0 N HNO_3 .

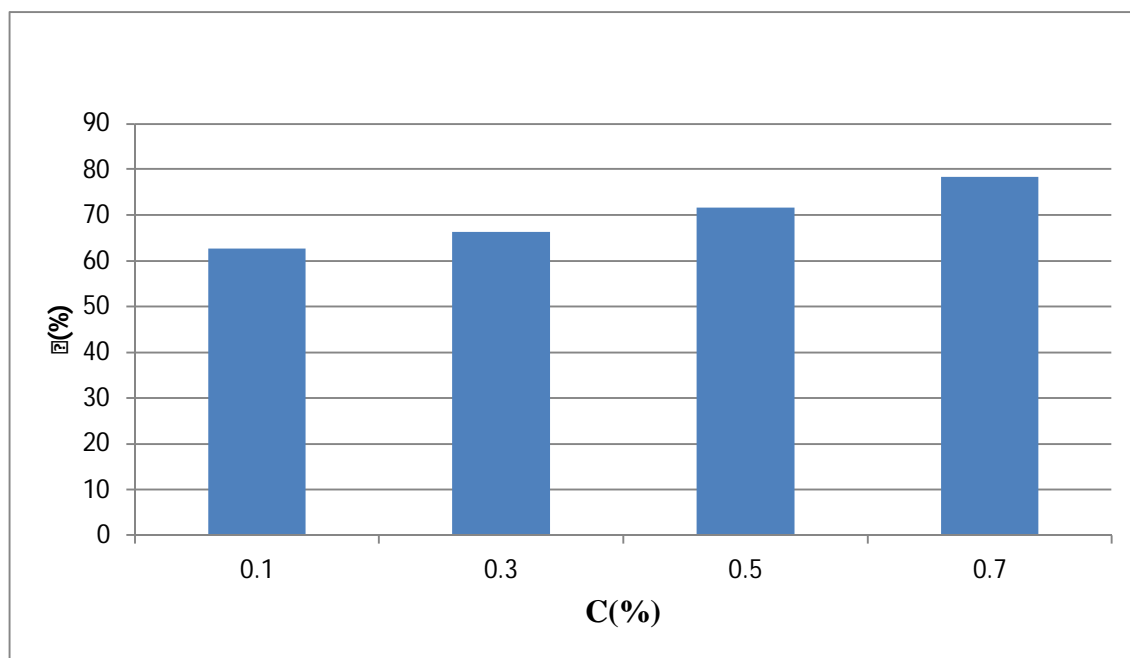


Fig. -4 : - Variation of percentage inhibition efficiency η (%) with inhibitor concentration C (%) for Aluminium in 2.0 N HNO_3

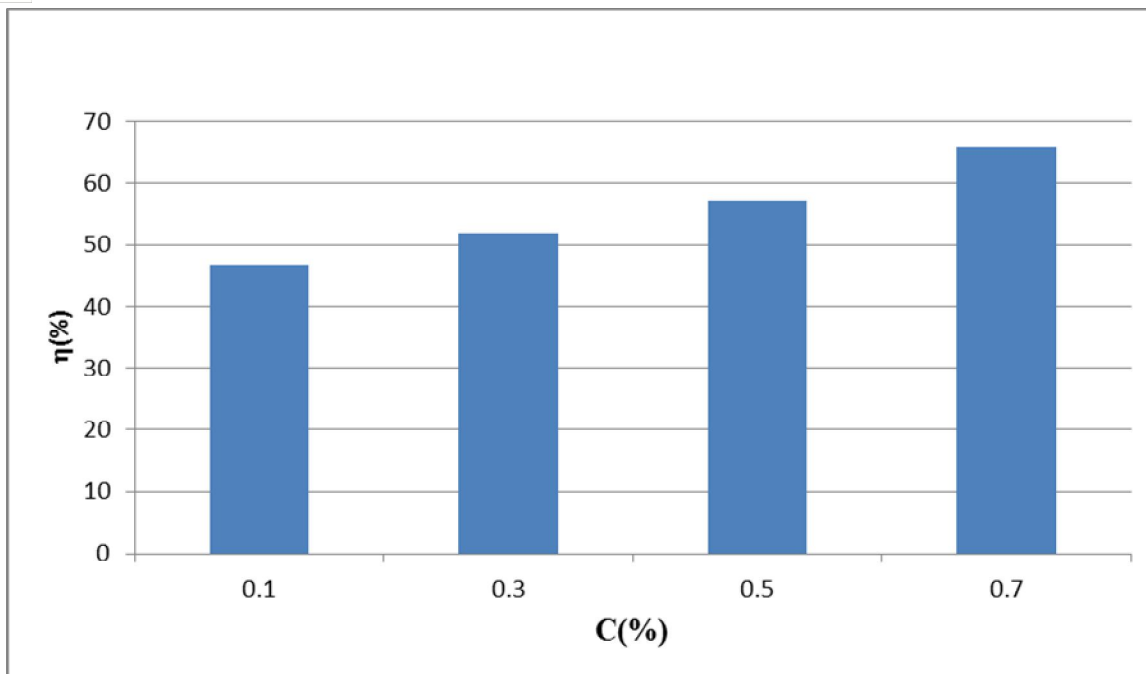


Fig. -5 : - Variation of percentage inhibition efficiency η (%) with inhibitor concentration C (%) for Aluminium in 0.1 N HNO_3 in presence of additive KNO_3 .

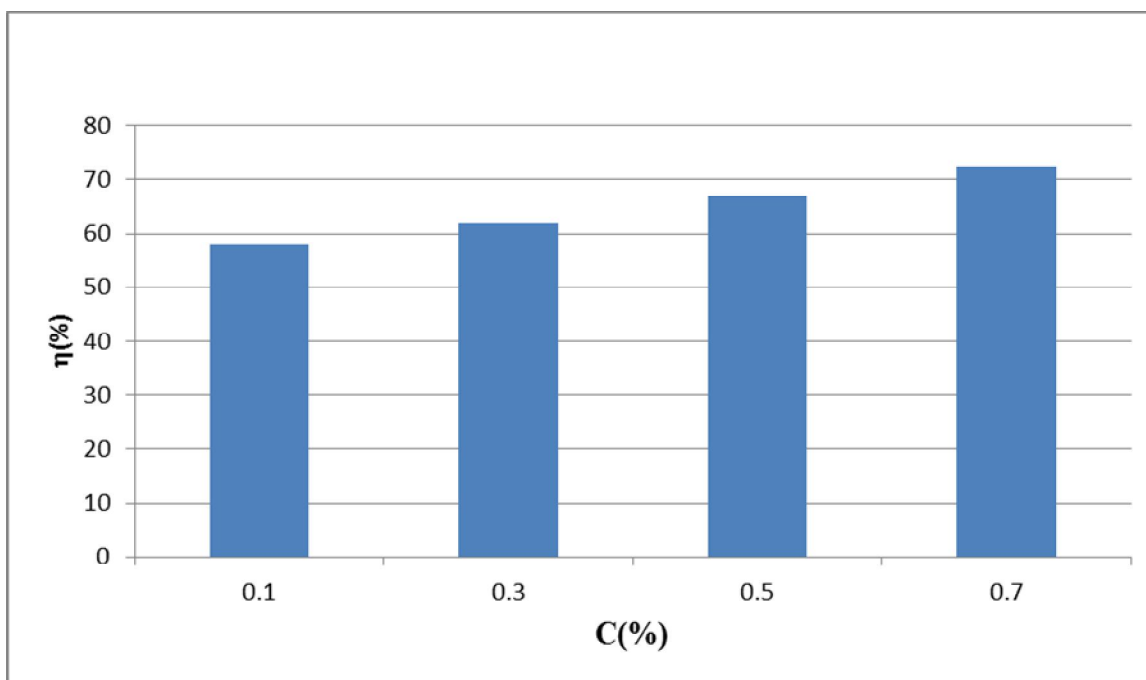


Fig. -6 : - Variation of percentage inhibition efficiency η (%) with inhibitor concentration C (%) for Aluminium in 0.5 N HNO_3 in presence of additive KNO_3 .

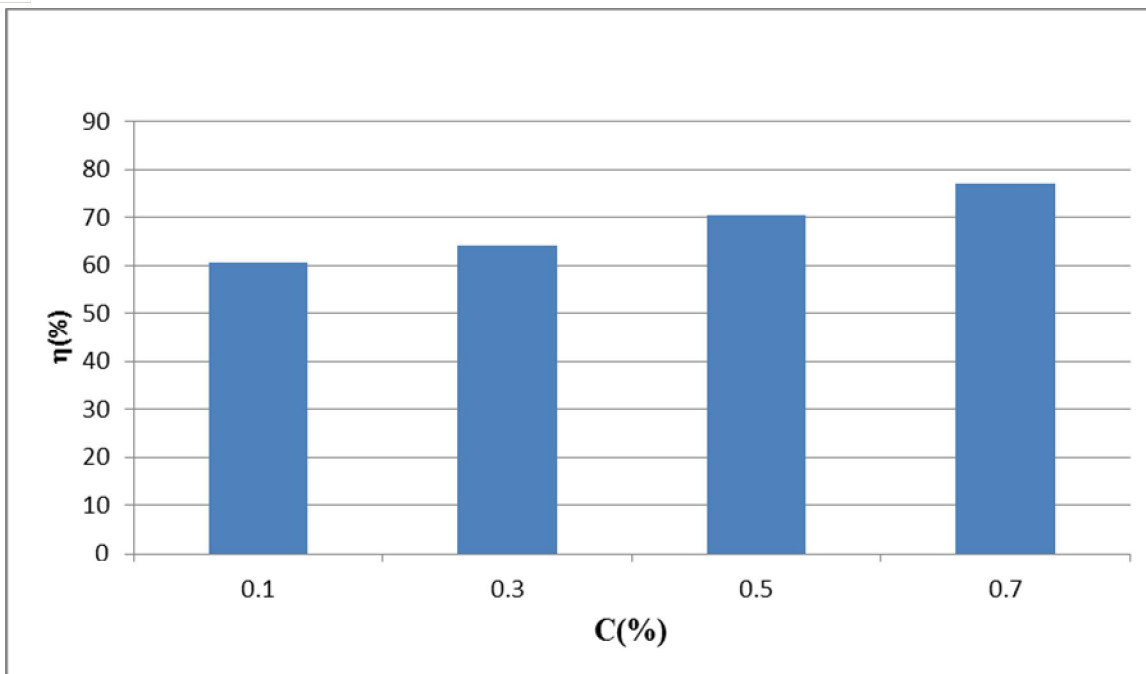


Fig. -7 : - Variation of percentage inhibition efficiency η (%) with inhibitor concentration C (%) for Aluminium in 1.0 N HNO_3 in presence of additive KNO_3 .

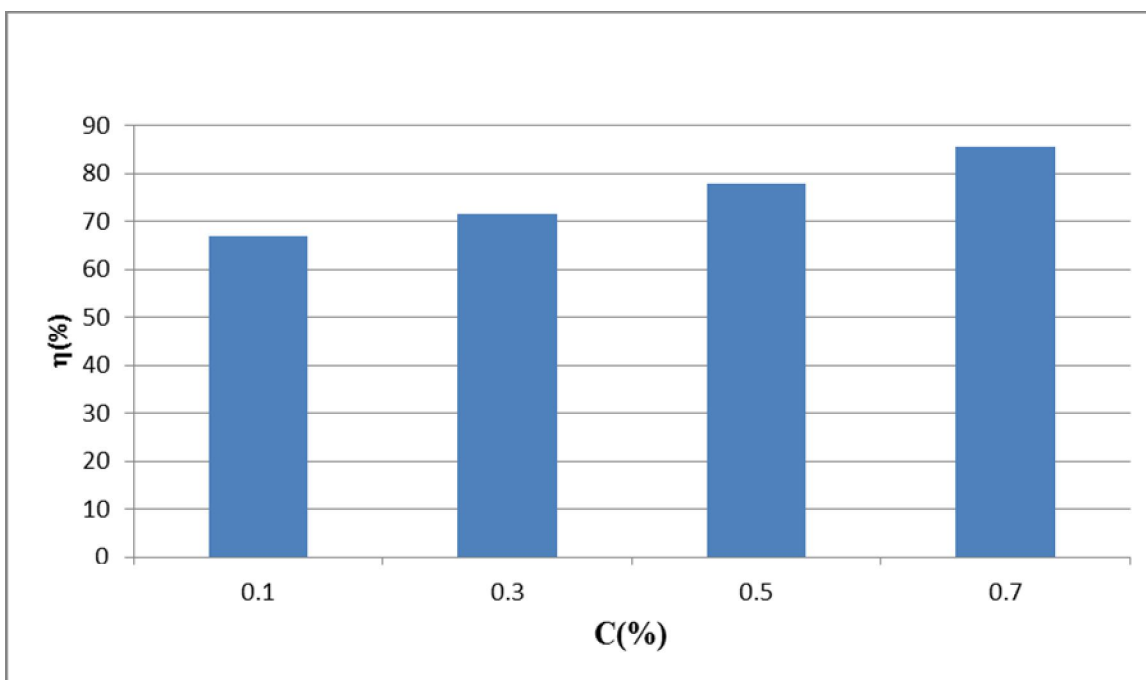


Fig. -8 : - Variation of percentage inhibition efficiency η (%) with inhibitor concentration C (%) for Aluminium in 2.0 N HNO_3 in presence of additive KNO_3 .

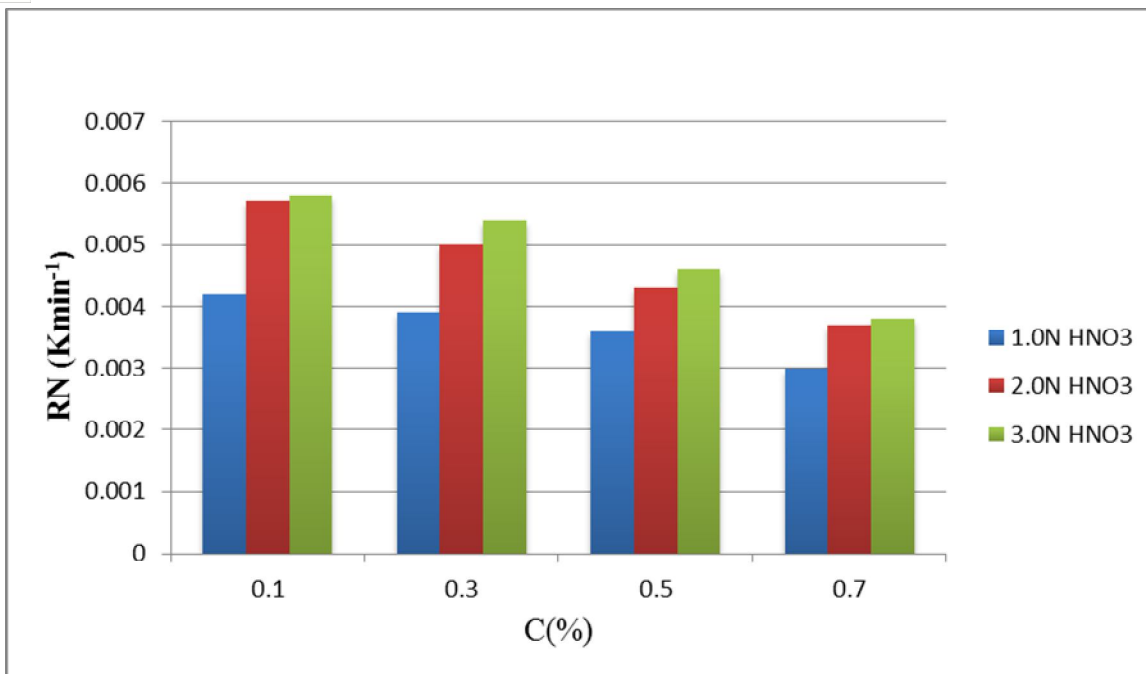


Fig. -9 : - Variation of Reaction Number with inhibitor concentration C (%) for Aluminium in HNO₃.

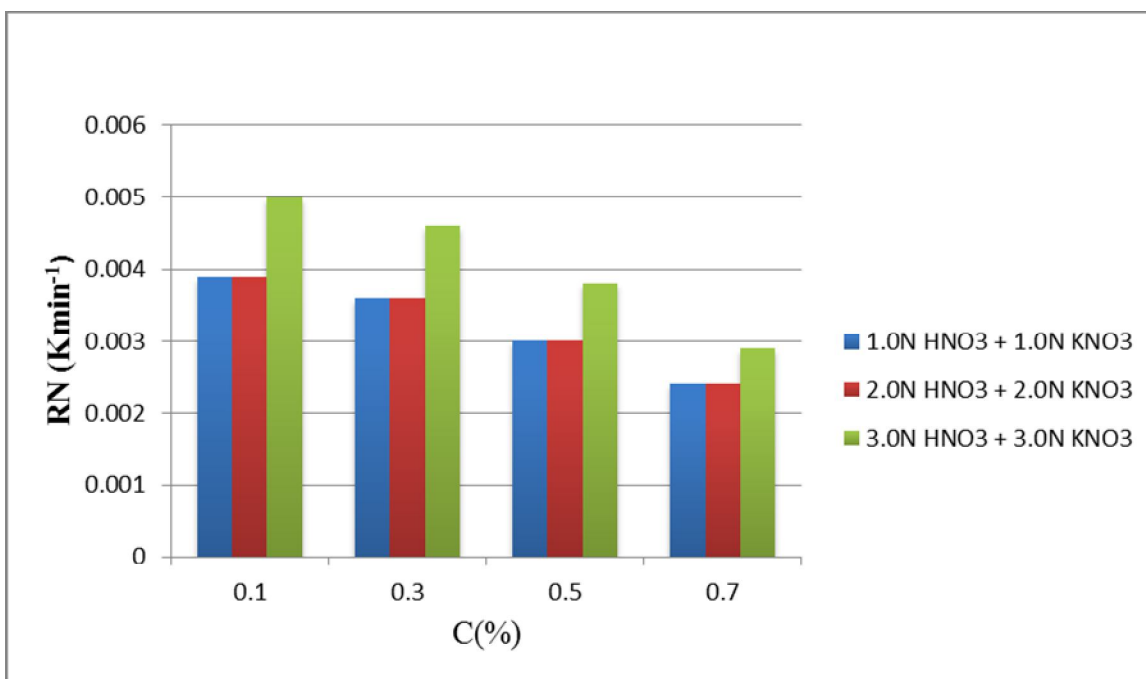


Fig. -10 : - Variation of Reaction Number with inhibitor concentration C (%) for Aluminium in HNO₃ in presence of additive KNO₃.



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