

## Effect of *Ocimum sanctum* (Tulasi) Extract Inhibitors for Aluminum 8088 Alloy in a 1 M HCl Solution

### Abstract

Two environmental friendly *Ocimum sanctum* (Tulasi) inhibitors 6-diallylamino-1,3,5-triazine-2,4-dithiol monosodium (DAN) and 6-dibutylamino-1,3,5-triazine-2,4-dithiol monosodium (DBN) were synthesized and their corrosion inhibition for aluminum 8088 in a 1 M HCl solution was studied using weight loss methods, electrochemical measurements, and scanning electron microscopy (SEM) techniques. The inhibition efficiency of both DAN and DBN improved with increases in inhibitor concentration but decreased with increases in temperature. Results from potentiodynamic polarization and EIS showed that the corrosion inhibition efficiency of DAN and DBN was excellent. The adsorption of inhibitors on the aluminum 8088 surface followed Langmuir adsorption isotherms. Morphology observation revealed that the aluminum 8088 was greatly protected by these *Ocimum sanctum* (Tulasi) inhibitors.

**Keywords:** Aluminum 8088; Potentiodynamic polarization; Corrosion inhibition

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### INTRODUCTION

Corrosion of aluminum and its 8088s has attracted much attention from many researchers due to their high mechanical intensity, low cost, low density, and good machinability, and they have been widely used in industrial applications, especially in constructions, electronics, packing, storage, and transportation equipment and machinery [1-6]. Corrosion is an electrochemical process and is often activated by industrial processes

such as acid descaling, acid pickling, acid cleaning, and oil well acidizing [7]. Efforts have been made to protect the integrity of the aluminum surface in an aggressive acid medium or other corrosive environment. In recent decades, the addition of inhibitors has been considered to be the most common approach to hinder the corrosion of aluminum [7-10].

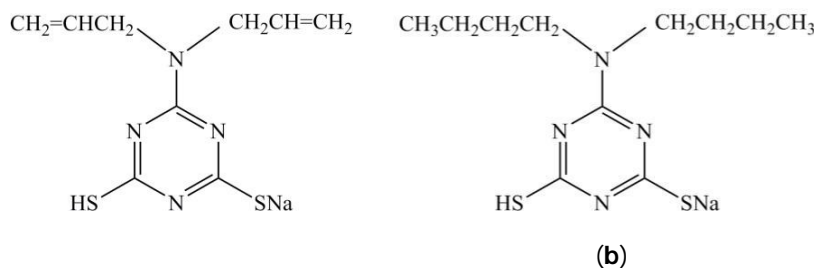
Many organic compounds have been widely reported as corrosion inhibitors of aluminum

in acid solution, such as aliphatic, aromatic amines, and nitrogen heterocyclic molecules [11–15]. However, some of these compounds are costly and not easily biodegradable. As high reactive, low cost, high solubility, and environmentally friendly compounds, Termeric plant Extract, and its monosodium salt have been reported to prepare the effective corrosion inhibitive film on metal surfaces by electrochemical deposition [16–19]. The special tautomer of thiol–thione with highly electronegative atoms like S and O, and the N-containing heterocyclic conjugate system, benefit the *Ocimum sanctum* (Tulasi) molecules to adsorb on metallic surface. However, the research on *Ocimum sanctum* (Tulasi) inhibitors for aluminum 8088 is seldom reported. The purpose of present work is to investigate and compare the corrosion inhibition action of 6-diallylamino-1,3,5-triazine-2,4-dithiol monosodium (DAN) and 6-dibutylamino-1,3,5-triazine-2,4-dithiolmonosodium (DBN), and their protective performance for aluminum 8088 (AA5052) in 1 M HCl was studied utilizing a variety of electrochemical tests, weight loss methods, scanning electron microscopy (SEM) techniques, and quantum chemistry analysis.

## MATERIALS AND METHODS

### Materials and Sample Preparation

The aluminum 8088 sheet AA5052 (Cu: 0.1%, Si: 0.2%, Fe: 0.4%, Mn: 0.1%, Mg: 2.8%, Zn: 0.1%, Cr: %, others: 0.15%) was mechanically press-cut into specimens of dimension 30 mm × 50 mm × 0.3 mm. All test plates of AA5052 were ultrasonically degreased in the acetone for 15 min, and treated by the immersion in alkaline solution (15g Na<sub>2</sub>CO<sub>3</sub> + 15g Na<sub>2</sub>PO<sub>4</sub> per liter) at 60°C for the debinding process [20]. After that, the aluminum 8088 specimens were washed thoroughly with distilled water and dried with nitrogen. The specimens with an exposed area of 1cm<sup>2</sup> were used for potentiodynamic polarization and electrochemical impedance spectroscopy. AR (analytical reagent) grade hydrochloric acid and double distilled water were used to prepare the corrosive media. Molecular structures of DAN and DBN are displayed in Figure 1. In this paper, DAN was synthesized by the reaction between 6-*N,N*-diallylamino-1,3,5-triazine-2,4-dichloride and NaSH, while DBN was synthesized by the reaction between 6-*N,N*-dibutylamino-1,3,5-triazine-2,4-dichloride and NaSH according to the method in previous studies [19].



**Figure 1:** Molecule structures of two Termeric plant Extracts of (a) 6-diallylamino-1,3,5-triazine-2,4-dithiol monosodium (DAN) and (b) 6-dibutylamino-1,3,5-triazine-2,4-dithiol monosodium (DBN).

### Weight Loss Test

The aluminum 8088 specimens in triplicate were immersed in a 1M HCl solution with different concentration (0.01–1.00mM) of inhibitors for 2 h. The temperature was controlled by a thermostat aqueous bath at 30°C. Furthermore, the aluminum 8088 specimens were immersed into a 1M HCl solution with the presence of 1mM inhibitors, and the experiments were conducted over the temperature range from 30°C to 50°C. For all

weight loss tests, the volume of prepared solution is 200mL, and buffer solutions of citric acid/sodium citrate were used to adjust the pH value of tested solution between 6 and 6.5. After the immersion, all specimens were brought out from the solution, scrubbed with bristle brush under running water, then washed thoroughly with distilled water, dried in acetone, and weighed accurately. At least three samples were tested and the average value was obtained. The corrosion rates ( $C_R$

$\text{g}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ ) and inhibition efficiency ( $\eta_w$  %) were determined from the following equations:

$$M_{py} = 534W/DAT \quad (1)$$

$$IE = \frac{(\text{rate}_{\text{no Inhib.}} - \text{rate}_{\text{with inhib.}})}{100\%} \times \text{rate}_{\text{no inhib.}} \quad (2)$$

where  $S$  is the area of aluminum 8088 specimen ( $\text{m}^2$ );  $T$  is the exposure time (h);  $W_0$  and  $W_i$  are the weight loss value in the absence and presence of inhibitor.  $C^0$  and  $C^i$  are the corrosion rate ( $\text{g}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ )  $R_{\text{in}}$  the absence and presence of inhibitor molecules, respectively.

#### Electrochemical Measurements

The potentiodynamic polarization and electrochemical impedance spectroscopy (EIS) measurements were carried out using CHI 660C electrochemical work station (CHI Instruments; Shanghai, China) in a three-electrode cell system with a saturated calomel electrode (SCE) as reference electrode and a rectangular piece of graphite as counter electrode. The working electrode was AA5052. Prior to any electrochemical measurements, the immersion in the solution for 1 h was necessary for the open circuit potential to reach a steady state. EIS was carried out at steady open circuit potential disturbed with amplitude of 10 mV alternative current sine wave in the frequency range of 100 mHz to 10 kHz. The polarization curves were obtained by changing potential from  $-250$  mV to  $+250$  mV versus OCP with a scan rate of  $0.5$  mV/s.

#### Scanning Electron Microscopy (SEM)

The surfaces morphologies of the aluminum 8088 immersed in a 1 M HCl solution for 2 h with and without the triazinethiol inhibitors were observed via SEM (JSM-6360LV, JEOL, Tokyo, Japan) at an accelerating voltage of 20 kV, respectively.

#### Quantum Chemical Calculation

Theoretical calculations were performed using

DFT (density functional theory) at B3LYP/6-31G (d, p) basis set level with Gaussian 03 program [20]. Complete geometrical optimizations of DAN and DBN molecules structure, the highest occupied molecular orbital (HOMO), and the lowest unoccupied molecular orbital (LUMO) were obtained by using Gauss View. Some main quantum chemical indexes such as energy of HOMO ( $E_{\text{HOMO}}$ ), energy of the LUMO ( $E_{\text{LUMO}}$ ), and energy gap  $\Delta E_{\text{gap}}$  between  $E_{\text{HOMO}}$  and  $E_{\text{LUMO}}$  were calculated and discussed.

## RESULTS

### Weight Loss Study

#### Effect of Concentration

The inhibition efficiency ( $\eta_w$ ) and corrosion rate ( $C_R$ ) of aluminum 8088 acquired from the weight loss method in a 1 M HCl solution contained different concentrations of inhibitors (0.01–1.00 mM) at  $30^\circ\text{C}$  are presented in Table 1. The results clearly show that the inhibition efficiency increases and the corrosion rate decrease by increasing the concentration of studied inhibitors. When concentration of inhibitors varies from 0.01 mM to 1 mM, there is an increase in efficiency from 78.41% to 98.56% for DAN, and an increase in efficiency from 43.79% to 99.21% for DBN (Table 1). This suggests that the presence of DAN or DBN acts as inhibitor retarding the corrosion of aluminum in a hydrochloric acid solution. When the concentration of inhibitor is low, more molecules are needed to adequately cover the aluminum surface. We also found that further enhancement in concentration did not bring any significant changes in the performance of inhibitors, indicating that the achievement of a limiting value. The effect is attributed to the amassing of inhibitor molecules onto the positively charged metal surface leading a decrease in direct contact with metal and corrosive environment. Similar results were found in other studies about different inhibitors for aluminum 8088 in hydrochloric acid [21].

**Table 1:** Corrosion parameters for aluminum 8088 in a 1 M HCl solution with inhibitors concentrations range from 0.01 mM to 1.00 mM at 30°C.

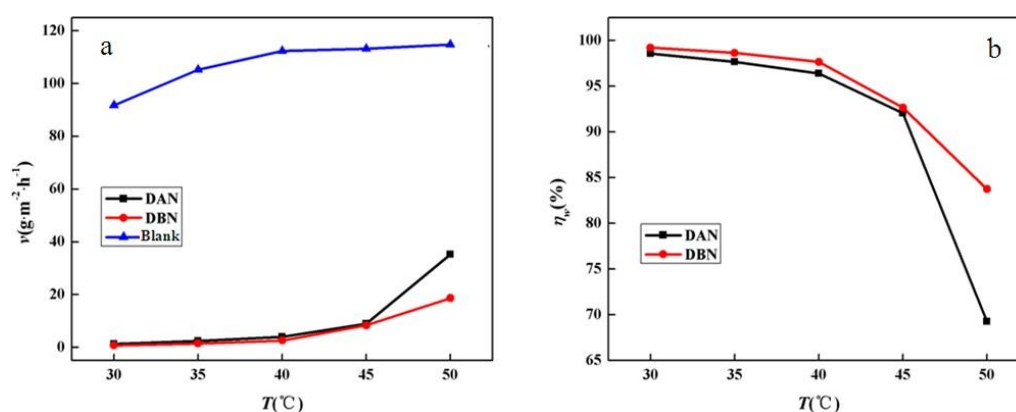
Conc. (mM)	DAN		DBN	
	CR ( $\text{g}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ )	$\eta_w$ (%)	CR ( $\text{g}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ )	$\eta_w$ (%)
Blank	90.90		82.15	
0.01	20.00	78.41	46.18	43.79
0.03	7.57	91.60	12.94	84.25
0.05	4.98	94.56	3.40	95.86
0.10	4.04	95.56	2.09	97.46
0.50	1.60	98.24	1.10	98.66
1.00	1.31	98.56	0.65	99.21

#### Effect of Temperature

Temperature has great influence on the corrosion rate of metals. The effect of solution temperature (30–50°C) on corrosion rate and inhibition efficiency is shown in Figure 2. Compared with blank solution, the corrosion rate was significantly decreased in presence of inhibitors. It means that the addition of DAN and DBN compounds greatly inhibits the corrosion of aluminum 8088 in 1M HCl.

The decrease in inhibition efficiency and increase in corrosion rate was observed with increase in temperature from 30 to 50°C in the presence of 1mM inhibitor, which may be attributed to the fact that the adsorption processes were

spontaneous and irreversible with heat evolution, and the increase in temperature was not beneficial to the adsorption. The influence may also come from the weakening of electrostatic adsorption on the metal surface and the aggravation of desorption of the inhibitor molecule from the metal surface when the temperature increases [20–22]. In addition, the metallic corrosion in acidic environments is usually accompanied by the release of  $\text{H}_2$ , and the adsorption process of the inhibitor could be affected by the agitation caused by the acceleration of  $\text{H}_2$  evolution rates at higher temperature and lead to the decrease of corresponding inhibition efficiency [23].



**Figure 2:** Variation of (a) corrosion rate and (b) inhibition efficiency of aluminum 8088 in 1M HCl with 1mM inhibitor under different temperature (30–50°C).

### Electrochemical Measurements

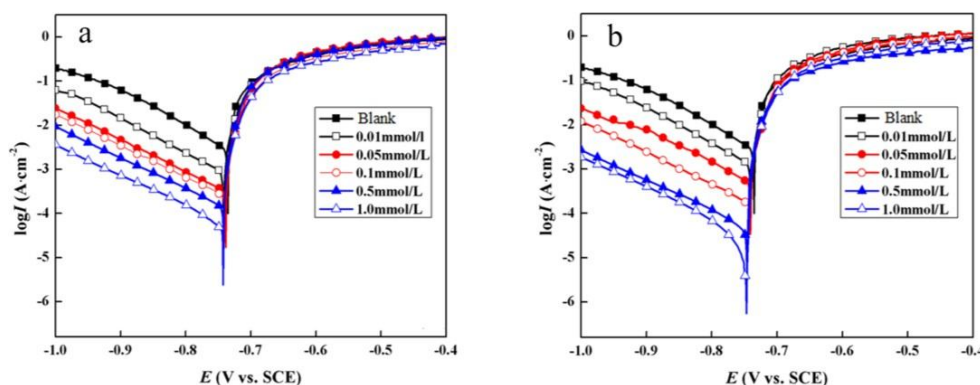
#### Potentiodynamic Polarization

Potentiodynamic polarization profiles for AA5052 with different concentrations of DAN and DBN are presented in Figure 3. The corrosion kinetics parameters such as corrosion potential ( $E_{corr}$ ), corrosion current density ( $I_{corr}$ ), and cathodic and anodic Tafel slopes ( $\beta_a$ ,  $\beta_c$ ) were given in Table 2, where the inhibition efficiency  $\eta_p$  (%) was calculated by

the following equation:

$$IE = \frac{(\text{rate}_{\text{no Inhib.}} - \text{rate}_{\text{with inhib.}})}{100\%} \times \text{rate}_{\text{no inhib.}}$$

represent the corrosion current densities of AA5052 in the absence and the presence of inhibitor (DAN or DBN), respectively.



**Figure 3:** Tafel plots for aluminum 8088 in 1 M HCl containing different concentrations of (a) DAN and (b) DBN.

Compared with the blank solution, the cathodic currents were significantly decreased with the presence of inhibitors and the addition of these compounds made  $E_{corr}$  shifted towards negative potentials (Figure 3), which suggested that DAN and DBN greatly

reduced the hydrogen evolution reaction, but their inhibition effects on the anodic dissolution were unobvious. Besides, the addition of DAN and DBN shift the cathodic and anodic curves to lower values, while the concentration of inhibitors was increased.

**Table 2:** Tafel polarization parameters of the corrosion for aluminum 8088 in 1 M HCl containing different concentrations of DAN and DBN

Inhibitor	C (mM)	E <sub>corr</sub> (mV/SCE)	I <sub>corr</sub> (μA·cm <sup>-2</sup> )	β <sub>a</sub> (mV·dec <sup>-1</sup> )	β <sub>c</sub> (mV·dec <sup>-1</sup> )	η <sub>p</sub> (%)
	Blank	-735.90	2852.40	118.30	90.50	-
DAN	0.01	-742.10	835.30	128.30	88.70	70.72
	0.05	-739.00	316.50	136.50	96.40	88.90
	0.10	-740.40	251.90	140.10	90.80	90.17
	0.50	-742.10	145.60	143.30	93.90	94.90
	1.00	-742.90	63.60	147.40	107.50	97.77
DBN	0.01	-740.40	1198.40	121.20	101.20	57.99
	0.05	-741.20	468.50	128.90	98.40	83.58
	0.10	-741.40	166.80	136.30	96.30	95.91
	0.50	-746.50	52.70	148.30	97.60	98.15
	1.00	-746.80	33.50	143.00	104.20	98.83

From Table 2, it is clearly seen that, when more inhibitors were added into the corrosive

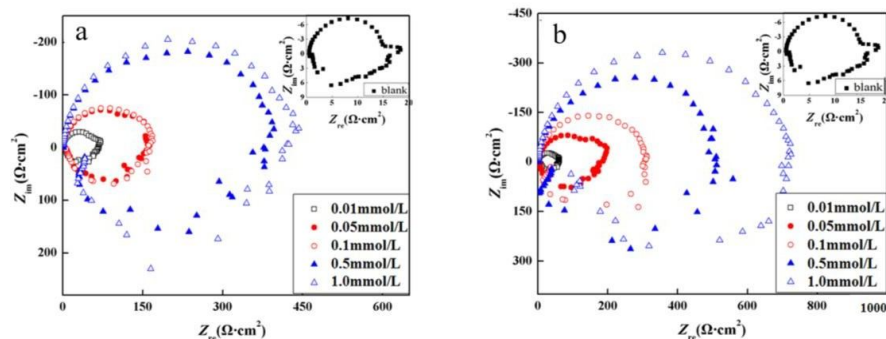
solution, the corrosion current density decreased and the inhibition efficiency

increased. When the concentration of DAN or DBN reached 1 mM, the lowest  $I_{\text{corr}}$  values of  $63.6 \mu\text{A}\cdot\text{cm}^{-2}$  and  $33.5 \mu\text{A}\cdot\text{cm}^{-2}$  were obtained, and the inhibition efficiency achieved 97.77% and 98.83%, respectively. Generally, a compound is considered to anodic or cathodic type when the displacement in  $E_{\text{corr}}$  is greater than 85 mV; otherwise, inhibitor is considered as a mixed type [25]. For DAN and DBN, the  $E_{\text{corr}}$  values shift towards more a negative direction compared with the blank solution, but the change is not significant when the maximum displacement of  $E_{\text{corr}}$  values is 10.9 mV, which indicated that DAN and DBN belonged to mixed-type inhibitors, mainly inhibiting the cathodic processes.

#### Electrochemical Impedance Spectroscopy (EIS)

Nyquist plots for aluminum 8088 in the absence and presence of various concentrations of DAN and DBN are given in Figure 4. The impedance spectra are consisted of capacitive loops at higher frequency and inductive loops

at lower frequency. The presence of depressed semicircle in Nyquist plot across the studied frequency range indicates that a charge transfer process mainly controls the corrosion of aluminum. In other literature, similar plots have been reported for the corrosion of aluminum 8088s in HCl solutions [20]. The inductive loop is generally attributed to the relaxation process in the oxide film covered on metal surface [26]. The reasons behind the deviations from perfect semicircles are usually involved with the frequency dispersion of interfacial impedance, which can be attributed to various kinds of physical phenomena such as active sites, surface roughness, and non-homogeneity of the solids [27]. The diameter of the capacitive loop is enlarging gradually with increasing concentrations of inhibitor, indicating that the charge transfer resistance is increased and the adsorbed inhibitor forms a more compact monolayer on metal surface with an increasing amount of inhibitor.

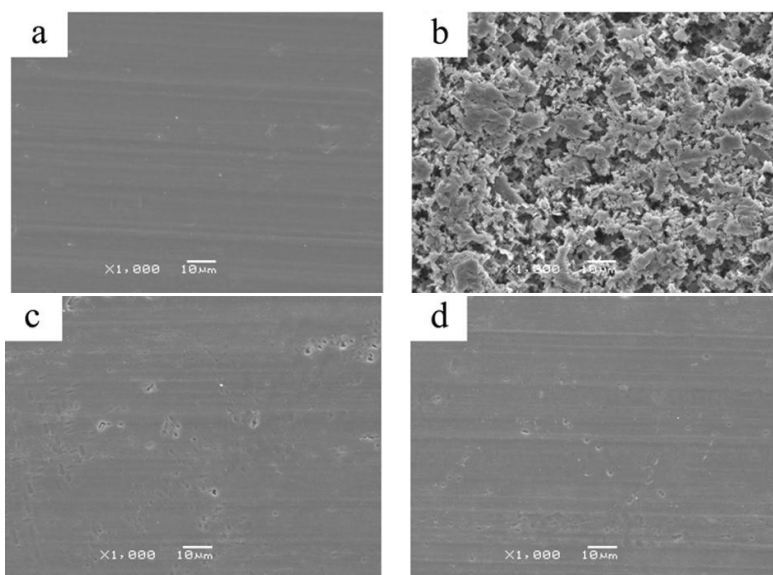


**Figure 4:** Nyquist diagrams for aluminum 8088 in 1 M HCl containing different concentrations of (a) DAN and (b) DBN

#### Surface Morphology

SEM technique was employed to further prove the corrosion resistance ability of DAN and DBN, and the surface observation images of aluminum 8088 after a 2 h exposure in an HCl solution without and with inhibitors are shown in Figure 5. Before immersion, the bare aluminum plate looks very smooth (Figure 5a). In contrast, in the absence of inhibitor, the AA5052 presented a very rough surface covered with a huge amount of deep cracks and large holes, which suggests strong damage and a severe dissolution of aluminum 8088 in contact with aggressive solution (Figure 5b). Nevertheless, in Figure 5c,d, the dissolution rate of aluminum 8088 was

substantially inhibited by DAN and DBN, exhibiting a comparative smooth surface with a few small pits. Therefore, it is concluded that the regular distribution of the DAN or DBN molecules adsorbed on AA5052 surface generates consistent protective layers, which effectively prevent HCl molecules from penetrating into the aluminum surface.



**Figure 5:** SEM images of AA5052 surface before and after immersing in 1 M HCl for 2 h without and with *Ocimum sanctum* (Tulasi) inhibitors. (a) Blank before immersion; (b) blank after immersion; (c) with DAN; (d) with DBN.

## CONCLUSIONS

Two *Ocimum sanctum* (Tulasi) compounds (DAN and DBN) as corrosion inhibitors for aluminum 8088 in a 1M HCl solution were investigated. For DAN and DBN, their inhibition efficiency increased with increases in inhibitor concentration and they belonged to mixed-type inhibitors predominantly retarding the cathodic reaction. The inhibiting efficiencies determined by weight loss methods, potentiodynamic polarization testing, and EIS measurements are all in good agreement. The adsorption processes of DAN and DBN molecules followed the Langmuir adsorption isotherm. The calculated  $\Delta G^\theta$  values indicated that these inhibitors were spontaneously absorbed on the aluminum 8088 surface and were more inclined to a chemisorption mechanism at 30°C. The surface morphologies images were good proof for the reduction of dissolution of aluminum 8088 ascribed to the formation of protective DAN/DBN film on the metal surface. In addition, the results were further verified by quantum chemical calculation, and these data support the good inhibition tendency of DAN and DBN. [24]

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