

## Corrosion Inhibition effect of Antimony Potassium Tartrate for Q235 Carbon Steel in NaCl Solutions

Shifang WANG, Yu ZUO, Yuming TANG, Xuhui ZHAO\*

Beijing Key Laboratory of Electrochemical Process and Technology for Materials, Beijing University of Chemical Technology, Beijing 100029, China

\*E-mail: [xhzhao@mail.buct.edu.cn](mailto:xhzhao@mail.buct.edu.cn)

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The inhibition effect of antimony potassium tartrate on the corrosion of Q235 Carbon Steel in NaCl solutions was studied through weight loss measurement, potentiodynamic polarization, scanning electron microscopy (SEM), and X-ray photoelectron spectroscopy (XPS). Thermodynamic calculations were carried out to explore the adsorption mechanism of antimony potassium tartrate. The results showed that antimony potassium tartrate can inhibit the corrosion of Q235 carbon steel in a 0.01M NaCl solution. A maximum inhibition efficiency of 63.5% was observed in the presence of 400 ppm of antimony potassium tartrate. In a 0.01M NaCl + 0.15 M NaHCO<sub>3</sub> solution, the inhibition efficiency increased drastically and reached 95.6% in the presence of 300 ppm of antimony potassium tartrate. Both chemical and physical adsorption occurred, and the Langmuir isotherm model was found to accurately describe the process.

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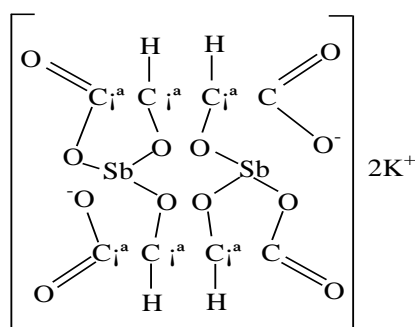
**Keywords:** Corrosion inhibition, Antimony potassium tartrate, Carbon steel, Adsorption

### 1. INTRODUCTION

The use of inhibitors is one of the most practical methods for the protection of metals against corrosion. The most effective and efficient inhibitors are organic compounds containing  $\pi$  bonds and heteroatoms (P, S, N, and O), and inorganic compounds, such as chromate, dichromate, and nitrite. Unfortunately, most of the inhibitors used are toxic, to different extents, to both humans and the environment. Thus, the development of novel non-toxic corrosion inhibitors of natural source has been considered to be important and desirable. Drugs (chemical medicines) are recognized as potential candidates to replace traditional toxic corrosion inhibitors due to their natural origin, non-toxic characteristics as well as negligible negative impact on the environment [1–8].

Some research has been carried out concerning the use of organic drugs as corrosion inhibitors

[9–21]. Morad et al. [9] studied the inhibition effect of cefatrexyl on iron corrosion in 1M solutions of HCl, HClO<sub>4</sub>, H<sub>2</sub>SO<sub>4</sub> or H<sub>3</sub>PO<sub>4</sub> solutions (1M for each) and found that cefatrexyl acts as a weak inhibitor in HCl solutions at 30°C, while it shows excellent inhibition performance in the remaining acids. The adsorption of cefatrexyl on iron surface obeys the Langmuir isotherm in an HCl solution, whereas in H<sub>2</sub>SO<sub>4</sub>, H<sub>3</sub>PO<sub>4</sub> and HClO<sub>4</sub> solutions it follows Temkin's isotherm. Tüken et al. [10] studied the effect of nicotinamide on iron corrosion in chloride solutions, while Quraishi et al. [11–14] studied the adsorption properties and corrosion inhibition effects of cefalexin, ceftriaxone, cefotaxime sodium and ceftobiprole on steel in HCl solutions. However, there is lack of studies focusing on the use of inorganic drugs as inhibitors. Antimony potassium tartrate (C<sub>8</sub>H<sub>4</sub>K<sub>2</sub>O<sub>12</sub>Sb<sub>2</sub>) is an inorganic drug for the treatment of schistosomiasis, and is also often used as a pigment. Antimony potassium tartrate can be ionized into [C<sub>8</sub>H<sub>4</sub>O<sub>12</sub>Sb<sub>2</sub>]<sup>2-</sup> and K<sup>+</sup>. Considering the presence of heteroatoms in [C<sub>8</sub>H<sub>4</sub>O<sub>12</sub>Sb<sub>2</sub>]<sup>2-</sup> and its adsorption characteristic on carbon steel surface, antimony potassium tartrate was considered as a viable inhibitor and its effects on the corrosion of Q235 Carbon Steel in NaCl solutions was studied.



**Figure 1.** Structure of antimony potassium tartrate

## 2. EXPERIMENTAL

The test material was Q235 carbon steel. Its composition is detailed in Table 1.

**Table 1.** Chemical composition of Q235 carbon steel

Element	C	Mn	P	S	Si
Content (wt%)	0.14–0.22	0.30–0.65	≤0.045	≤0.050	≤0.030

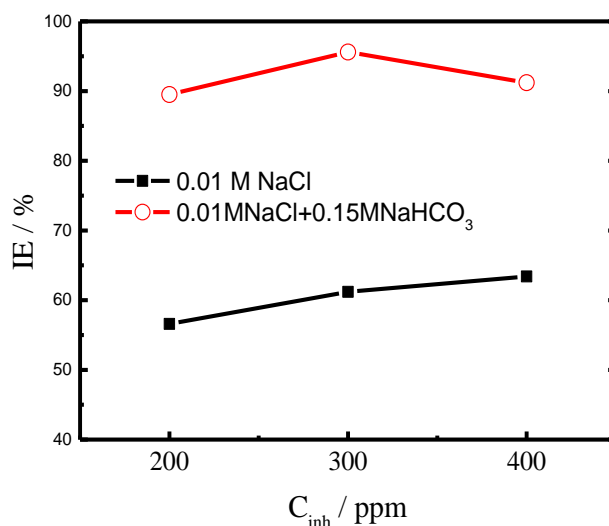
The samples were cut to the size of 50 mm×25 mm×2 mm, abraded with abrasive papers from 240# up to 1000#, then rinsed with deionized water and degreased with ethanol. The weight loss corrosion tests were carried out in following two solutions: 0.01 mol/L NaCl solution (pH=5.5) and 0.01 mol/L NaCl+0.15 mol/L NaHCO<sub>3</sub> solution (pH=11), each containing different concentrations of antimony potassium tartrate (APT). Test time was 30 days. Each experiment was repeated at least three

times. All the reagents used in this study were of analytic grade, and were purchased by Sinopharm Chemical Reagent Co., Ltd, China.

Polarization measurements were performed using a CS350 electrochemical workstation (Wuhan CorrTest Instruments Corp. Ltd., China) with a typical three-electrode system. An Ag/AgCl electrode was used for reference, the counter electrode was made of Pt and the specimen served as the working electrode, with an exposed area of  $1\text{ cm}^2$ . The potentiodynamic polarization tests were carried out at a scanning rate of  $0.33\text{ mV/s}$  after immersion of the sample in the test solutions at room temperature for 30 min. The test solutions used in polarization tests were the same as those of used in the weight loss corrosion tests. XPS spectra of the sample surface were analyzed with an ESCALAB 250 (Thermo Fisher Scientific, USA) X-rays photoelectron spectrometer with a monochromated Al Ka source ( $1486.6\text{ eV}$ ). The operating pressure was less than  $10^{-8}\text{ Pa}$ . The measured binding energy (BE) range for the wide scan spectrum was from 0 to  $1350\text{ eV}$  in  $1\text{ eV}$  steps without sputtering. The binding energy values were calibrated by the C1s peak at  $285\text{ eV}$ . The data were fitted by XPSPEAK 4.1 software.

### 3. RESULTS AND DISCUSSION

#### 3.1 Weight-loss measurements



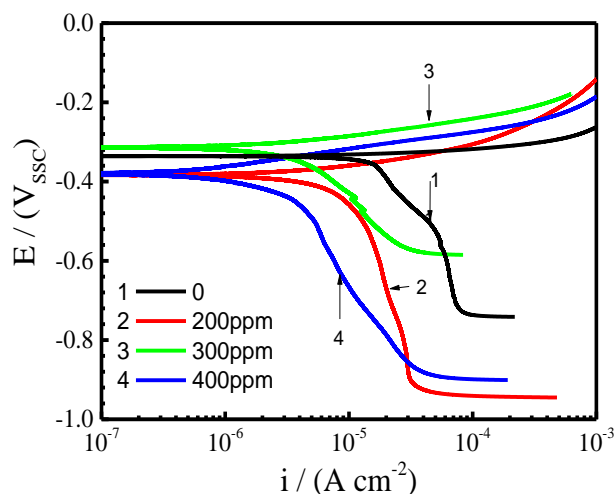
**Figure 2.** Results of weight lost corrosion test of carbon steel in two solutions with different concentrations of antimony potassium tartrate (APT)

Figure 2 shows the results of the weight loss corrosion tests of carbon steel in the  $0.01\text{ mol/L}$  NaCl solution and the  $0.01\text{ mol/L}$  NaCl+ $0.15\text{ mol/L}$  NaHCO<sub>3</sub> solution containing different concentration of APT after immersion for 30 days. For the first solution, as the APT concentration increased, the inhibiting efficiency also increased. The corrosion inhibition efficiency was about 63.5% when the APT concentration was 400 ppm. However, in the  $0.01\text{ mol/L}$  NaCl+ $0.15\text{ mol/L}$  NaHCO<sub>3</sub>

solution, the inhibition efficiency of APT increased noticeably, reaching a value of 95.6% for an APT concentration of 300 ppm. This result indicates a synergistic effect between APT and NaHCO<sub>3</sub>, implying an APT synergistic effect. A further increase in APT concentration to 400 ppm caused a slight decrease in the inhibition efficiency.

### 3.2 Polarization curves

The polarization curves of carbon steel in the 0.01 mol/L NaCl solution containing different concentrations of APT are shown in Figure 3. The analysis results are shown in Table 2. From Figure 3 and table 2, it can be seen that carbon steel was in active state without adding APT inhibitor and the corrosion rates were relatively fast. After adding APT, the polarization behavior of steel in the test solution was almost the same with those without adding inhibitor. However, the corrosion current densities decreased obviously after addition of APT. the corrosion inhibitor (APT) had an obvious influence on the cathodic polarization of carbon steel, while the corrosion potential and anodic polarization curves changed only slightly. As the APT concentration increased, the corrosion currents decreased. This implies that APT inhibits the cathodic process of corrosion and is therefore a cathodic-type corrosion inhibitor [22]. The inhibition efficiency reached 83.3% for an APT concentration of 400 ppm.

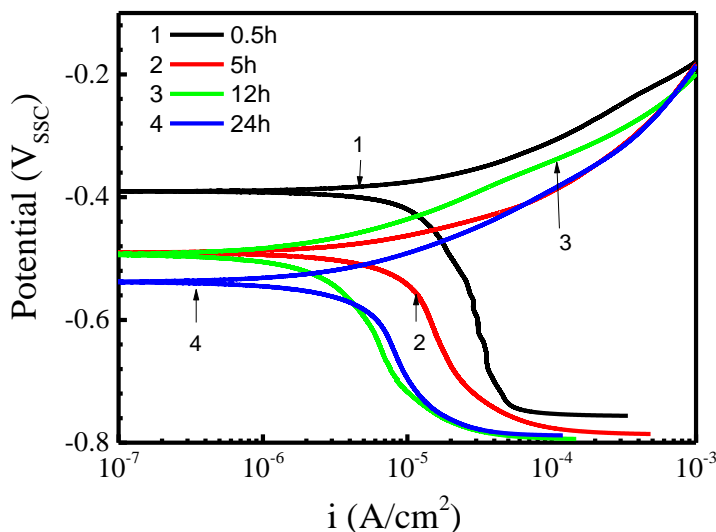


**Figure 3.** Polarization curve of carbon steel in the NaCl solution with different concentrations of APT

**Table 2.** Results of the polarization test of carbon steel in the 0.01 mol/L NaCl solution containing different concentrations of APT

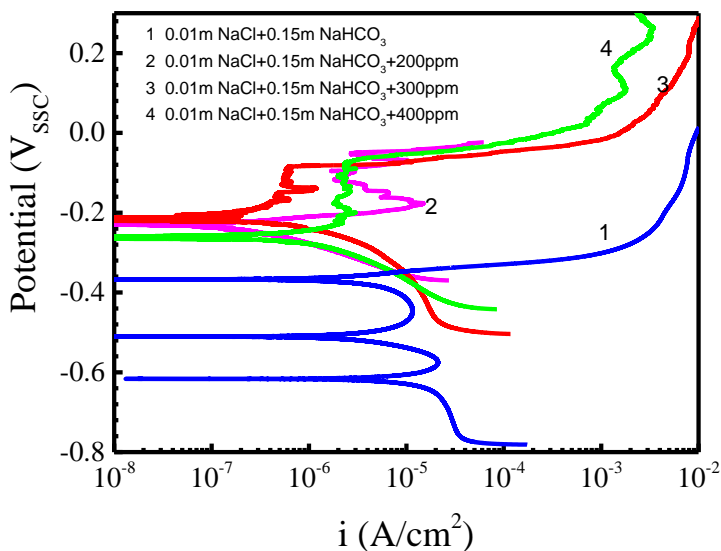
APT Concentration (ppm)	E <sub>corr</sub> ( mv )	I <sub>corr</sub> ( μA/cm <sup>2</sup> )	IE(%)
0	-333.11	15.98	-
200	-392.08	6.15	61.5
300	-315.59	3.20	80.0
400	-380.29	2.67	83.3

In order to further study the influence of immersion time on the corrosion behavior of carbon steel in the 0.01 mol/L NaCl solution, polarization measurements were performed after different immersion times. The polarization curves of carbon steel in the 0.01 mol/L NaCl solution containing 400 ppm of APT after immersion of the sample for 0.5, 5, 12, and 24 h are shown in Figure 4. The polarization behavior of steel in the test solution was almost with immersion time. However, as the immersion time was prolonged, the corrosion potential gradually shifted in the negative direction, showing the cathodic inhibitive effect of APT.



**Figure 4.** Polarization curves of carbon steel in the NaCl solution containing 400 ppm APT after immersion for different times

Figure 5 shows the polarization curves of carbon steel in the 0.01 mol/L NaCl +0.15 mol/L NaHCO<sub>3</sub> solution containing different concentrations of APT after immersion for 1 h. The analysis results are shown in Table 3.



**Figure 5.** Polarization curves of carbon steel in the NaCl +NaHCO<sub>3</sub> solution with different concentrations of APT

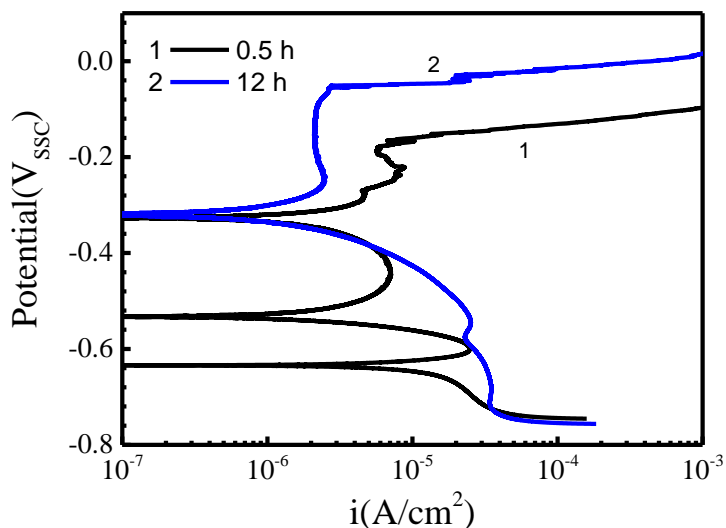
From Figure 5, it can be seen that there are two peaks in the polarization curves of carbon steel in the 0.01 mol/L NaCl +0.15 mol/L NaHCO<sub>3</sub> solution, showing the presence of an unstable passive state. When adding APT, the corrosion potential shifted in the positive direction and a stable passivation region was formed, implying that the APT inhibitor can promote the passivation and decrease the passive current density. Zhou et al [23] studied the compounded inhibition of sodium molybdate and benzotriazole on pitting corrosion of Q235 steel in 0.01M NaCl +0.1M NaHCO<sub>3</sub> solution, and also pointed out that the compound inhibitors effectively promoted passivation of steel and inhibited pitting corrosion, particularly in basic solutions. Wang et al [24] also found that carbon steel was passivated in 0.1M NaCl + Ca(OH)<sub>2</sub> solution when the solution pH value increased to 9.5~10.5, the calcium lignosulfonate (CLS) can promote the passivation and effectively inhibited both the cathodic and the anodic reactions. A possible reason is that an FeCO<sub>3</sub> film can be formed on the surface of the carbon steel in the presence of the NaCl+NaHCO<sub>3</sub> solution, although the film is not compact [22]. On the other hand, in the NaCl+NaHCO<sub>3</sub> solution containing APT, the adsorption of APT improves the corrosion resistance of the FeCO<sub>3</sub> film. In addition, when adding 400 ppm of APT, the corrosion current of carbon steel increased. This result is consistent with the weight loss tests. According to the fitting results of Table 3, in the NaCl+NaHCO<sub>3</sub> solution, APT shows a good inhibition effect on carbon steel, and the inhibitive efficiency reaches 97.5%.

**Table 3.** Fitting results of polarization after adding APT to the NaCl +NaHCO<sub>3</sub> solution

APT Concentration (ppm)	E <sub>corr</sub> ( mv )	I <sub>corr</sub> ( μA/cm <sup>2</sup> )	IE(%)
0	-617.7	3.53	-
200	-238.5	0.16	95.5
300	-213.7	0.09	97.5
400	-254.0	0.53	85.0

The polarization curves of carbon steel in the NaCl+NaHCO<sub>3</sub> solution with APT, after immersion for different times, are shown in Figure 6. For an immersion time of 0.5 h, two peaks are visible in the polarization curve, indicating the presence of an unstable passive state. This result indicates that the adsorption film of APT on the surface of the steel substrate is incomplete. When prolonging the immersion time to 12 h a stable passive region in the polarization curves could be observed, indicating the adsorption and film formation processes of the inhibitors on the surface. The inhibition efficiency of APT in the 0.01 mol./L NaCl+0.15 mol/LNaHCO<sub>3</sub> solution increased with immersion time, consistent with the results obtained in the NaCl solution.

According to above results, it can be seen that the inhibition effect of APT on carbon steel in 0.01 mol./L NaCl+0.15 mol/LNaHCO<sub>3</sub> solution is better than that in 0.01 mol./L NaCl solution. It should be noted that the pH value of NaCl+NaHCO<sub>3</sub> solution increased because of adding NaHCO<sub>3</sub>, both OH<sup>-</sup> ions and APT can promote the passivation of carbon steel. However, adsorption of OH<sup>-</sup> and tartarate ions and interaction with steel surface is not clear and further research is needed.



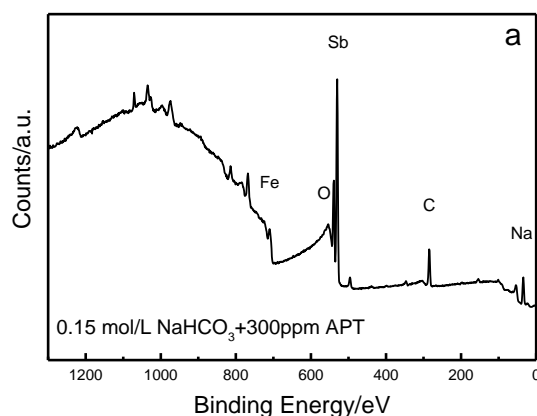
**Figure 6.** Polarization curves of carbon steel in the NaCl+NaHCO<sub>3</sub> solution containing 300 ppm APT after immersion for different times

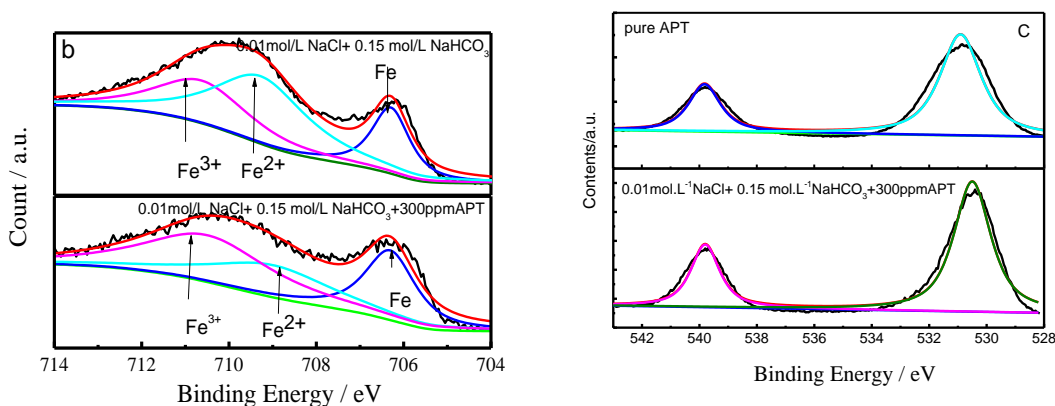
### 3.3 XPS results

The XPS analysis was carried out to study the surface composition of the samples immersed in the NaCl+NaHCO<sub>3</sub> solution with 300 ppm APT after 12 h. After the immersion period, the surface of the samples was rinsed and dried at room temperature. The obtained binding energy values were calibrated according to the binding energy of the C1s peak at 285 eV.

Figure 7a shows the wide-scan XPS spectra of the carbon steel sample. The XPS results show the presence of C, O, Fe and Sb. For the sample immersed in the solution containing APT, the peaks relative to Sb were easily recognizable, confirming the adsorption of APT.

Figure 7b presents the Fe 2p<sub>3/2</sub> spectra of the samples. As shown in the Figure, the Fe 2p<sub>3/2</sub> spectrum can be decomposed into two peaks, one attributable to Fe<sup>3+</sup> (710.5-711.5 eV) and Fe<sup>2+</sup> (709.5 eV), and the other peak to Fe<sup>0</sup> (706.7 eV) [25]. It can also be seen that the Fe<sup>3+</sup> content increased after adding APT, indicating that APT can promote passivation.





**Figure 7.** Wide-scan XPS spectra (a) of sample and High-resolution XPS spectra of Fe (b) and Sb(c) in the NaCl+NaHCO<sub>3</sub> solution with 300 ppm of APT after immersion for 12 h

Figure 7c shows the Sb 3d spectra of the samples, from which it can be seen that the peak positions of the binding energy of Sb onto the sample surface are almost the same as those relative to pure APT.

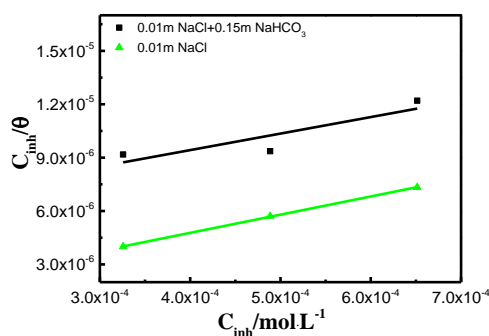
### 3.4 Adsorption mechanism

Physisorption and chemisorption are the two main types of adsorption of inhibitors on the surface of metals [26,27]. Some basic information on the interaction between the inhibitors and the metal surface can be obtained by the adsorption isotherm [28,29]. The surface coverage,  $\theta$ , of the samples immersed in the NaCl solutions with different concentrations of APT were calculated through the results of the weight-loss measurements, as shown in Figure 8. It is seen that the Langmuir adsorption isotherm fits the results adequately.

The Langmuir adsorption isotherm is expressed by the following equation:

$$\frac{\theta}{1-\theta} = K C_{inh} \tag{1}$$

where,  $C_{inh}$  is the concentration of APT and  $K$  is the equilibrium constant for the adsorption/desorption process.



**Figure 8.** Langmuir adsorption plots for carbon steel in NaCl solutions with different concentrations of APT



The standard free energy of adsorption ( $\Delta G_{\text{ads}}$ ) is calculated using the following equation [30]:

$$K = \frac{1}{55.5} \exp\left(\frac{-\Delta G_{\text{ads}}}{RT}\right) \quad (2)$$

where R is the gas constant and T the absolute temperature.

The  $\Delta G_{\text{ads}}$  values calculated for APT according to Eq.(2) are shown in Table 4. The negative value of  $\Delta G_{\text{ads}}$  suggests that APT is spontaneously adsorbed on the carbon steel surface. A previous study had reported that  $\Delta G_{\text{ads}}$  values more negative than  $-40$  kJ/mol can be considered to indicate the occurrence of chemisorption with the formation of a new coordinate type of bond or charge sharing [29].  $\Delta G_{\text{ads}}$  values of approximately  $-20$  kJ/mol or above may be considered to indicate physisorption [31] and electrostatic attraction [29]. The  $\Delta G_{\text{ads}}$  value in this test suggests that both physisorption and chemisorption may occur [32,33].

**Table 4.** Calculated thermodynamic parameters of APT

Solution system		K( $\times 10^3$ L/mol)	$\Delta G_{\text{ads}}$ (KJ/mol)
0.01 mol./L NaCl+	200ppm APT	4.066	-24.82
	300ppm APT	3.106	-24.16
	400ppm APT	2.649	-23.76
0.01 mol./L NaCl+0.15 mol/LNaHCO <sub>3</sub> +	200ppm APT	26.147	-29.43
	300ppm APT	44.432	-30.75
	400ppm APT	15.920	-22.50

#### 4. CONCLUSIONS

(1)Antimony potassium tartrate can, to some extent, inhibit the corrosion of carbon steel in a 0.01mol/L NaCl solution by inhibiting its cathodic process.

(2)In a 0.01 mol/L NaCl+0.15 mol/L NaHCO<sub>3</sub> solution, antimony potassium tartrate can promote the passivation of carbon steel and shows a good inhibition effect on the corrosion, acting as a mixed-type inhibitor. When the APT concentration is 300 ppm, the corrosion inhibition efficiency reaches 95.6%.

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