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Takahashi, K.; Bardwell, J. A.; MacDougall, B.; Graham, M. J.

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MECHANISM OF ANODIC DISSOLUTION AND PASSIVATION OF IRON—II. COMPARISON OF THE BEHAVIOR IN NEUTRAL BENZOATE AND ACETATE BUFFER SOLUTIONS

K. TAKAHASHI,* J. A. BARDWELL, B. MACDOUGALL and M. J. GRAHAM

Institute for Microstructural Sciences, National Research Council of Canada, Ottawa, Canada K1A 0R9

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Abstract—The kinetics of the dissolution and passivation of Fe has been studied in neutral benzoic acid/sodium benzoate buffer solution as a function of the total benzoate buffer concentration and pH. The results have been obtained from potential sweep experiments at 5 mV s^{-1} , which corresponds to quasi steady-state conditions. Two anodic peaks were found in the active dissolution region of the anodic voltammogram. A reaction order of about -1 with respect to benzoate concentration and of about 2 with respect to pH has been found for the Tafel region of the first peak. The corresponding reaction orders were -1 and 0.7 for the second peak. Galvanostatic cathodic reduction experiments were used to characterize the surface film formed during the anodic sweep. Anodic sweep experiments were also performed using mixtures of benzoate and acetate buffers. The behavior of Fe in benzoate and acetate buffer solutions has been compared and contrasted. Benzoate buffer is found to be a much better inhibitor than acetate buffer, and the results can be understood in terms of a strong interaction between the Fe surface and benzoate-containing adsorbates.

Key words: iron, dissolution mechanism, inhibitor, acetate, benzoate, passivation.

INTRODUCTION

It has long been recognized that benzoate and related ring-substituted compounds act as inhibitors for the Fe dissolution reaction[1–11]. In the preceding paper, the mechanism of Fe dissolution and passivation in neutral and slightly acidic sodium acetate/acetic acid buffer solutions has been investigated over a wide range of pH and buffer concentrations[12]. A model was proposed, which semi-quantitatively fitted the experimental anodic voltammograms, and yielded a simple, physical explanation for the observed reaction orders, with respect to pH and acetate buffer concentration. In this paper, similar experiments have been performed using the benzoate buffer system, and the results compared with those obtained in acetate. The previously proposed model is again used to give a semi-quantitative fit to the experimental data. Mixtures of acetate and benzoate have also been studied, and the inhibitive properties of the various solutions compared and contrasted.

EXPERIMENTAL

Experimental details are given in the preceding paper[12]. Benzoate buffer solutions were made by adjusting the pH of sodium benzoate (Anachemia) solutions with a saturated solution of benzoic acid (Aldrich, 99% + ACS reagent). Although small changes in the molar concentration are inevitable

with this procedure, the final concentrations obtained for all solutions in this study differed from the nominal concentrations by less than 5%, and frequently by less than 1%. In the discussion below, [benzoate] will refer to the total benzoate buffer concentration, $[\text{C}_6\text{H}_5\text{COOH}] + [\text{C}_6\text{H}_5\text{COO}^-]$. $[\text{C}_6\text{H}_5\text{COOH}]$ will be abbreviated [HB] and $[\text{C}_6\text{H}_5\text{COO}^-]$ as $[\text{B}^-]$. The ratio $[\text{HB}]/[\text{B}^-]$ will, of course, vary with pH. The concentration of the various species may be calculated from

$$[\text{HB}] = \frac{10^{-\text{pH}} [\text{benzoate}]}{K_a + 10^{-\text{pH}}}$$

$$[\text{B}^-] = [\text{benzoate}] - [\text{HB}],$$

where K_a is the equilibrium constant for the dissociation of HB and is given by

$$K_a = \frac{[\text{H}^+][\text{B}^-]}{[\text{HB}]} = 6.31 \times 10^{-5}.$$

Similarly, [HA], $[\text{A}^-]$ and [acetate] refer to the concentrations $[\text{CH}_3\text{COOH}]$, $[\text{CH}_3\text{COO}^-]$ and $[\text{CH}_3\text{COOH}] + [\text{CH}_3\text{COO}^-]$. Potentials were measured relative to a $\text{Hg}/\text{Hg}_2\text{SO}_4/0.15 \text{ N Na}_2\text{SO}_4$ reference electrode (0.421 V vs sce , 0.661 V vs she). All electrodes were galvanostatically reduced at $20 \mu\text{A cm}^{-2}$ for 300 s to remove the electropolish film before initiating a potential sweep at -1.5 V . A sweep rate of 5 mV s^{-1} was used in all experiments. This sweep rate is expected to yield only quasi steady-state data, as discussed in detail in the preceding paper[12]. Nevertheless, it does give valuable information on the trends with respect to solution concentration, pH and particularly regarding the

*Visiting scientist at NRC. Present address: Kurita Water Industries, Morinosato, Atsugi, 243-01, Japan.

Fig. 1. Effect of [benzoate] on anodic sweep voltammograms for Fe at 5 mV s^{-1} in pH 7 solution. Peak A' obtained at [benzoate] $\leq 0.5 \text{ M}$ is an artifact due to insufficient buffer capacity.

relative interactions of benzoate and acetate with the Fe surface.

RESULTS AND DISCUSSION

The effect of [benzoate] on the anodic sweep voltammograms of Fe at 5 mV s^{-1} is shown in Fig. 1. At [benzoate] $\geq 0.75 \text{ M}$, two anodic peaks are seen in the anodic dissolution region of the voltammogram, as in the case of acetate buffer. They are labeled A and B, in accord with previous work. At [benzoate] $\leq 0.5 \text{ M}$, an additional peak or shoulder, labeled A', appears. As previously determined for the acetate buffer system[12], this peak is an experimental artifact, due to cathodic polarization induced basicity at the electrode surface. The basicity results from a depletion of H^+ due to H_2 evolution during the cathodic polarization. It can be eliminated by providing sufficient buffer capacity in the solution, or by using a sufficiently slow sweep rate. Thus, meaningful results for the benzoate system can only be obtained for [benzoate] $\geq 0.75 \text{ M}$ in pH 7 solution at a sweep rate of 5 mV s^{-1} . From an examination of the Tafel regions of peaks A and B, the reaction orders with respect to [benzoate], $y_{A, [\text{benzoate}]}$ and $y_{B, [\text{benzoate}]}$, are clearly negative. Due to the small range of [benzoate] available, and because the linear Tafel regions are narrow, it was difficult to obtain an accurate value of the reaction orders, but both $y_{A, [\text{benzoate}]}$ and $y_{B, [\text{benzoate}]}$ are ca. -1 . The peak current in peak A ($j_{p,A}$) is

Fig. 2. Effect of pH on anodic sweep voltammograms for Fe at 5 mV s^{-1} in [benzoate] = 1 M solution.

nearly constant with [benzoate], while $j_{p,B}$ decreases with increasing [benzoate].

The effect of pH on the anodic sweep voltammogram of Fe at 5 mV s^{-1} is shown in Fig. 2. The range of pH which can be investigated is limited both by the buffer capacity and by the solubility of benzoic acid. Curves for $\text{pH} \geq 7.3$ show some indication of polarization induced basicity; thus meaningful data is restricted to $\text{pH} \leq 7.2$. Nevertheless, the reaction orders in the Tafel region of both peaks A and B are positive, and have values of $y_{A, \text{pH}} \approx 2$ and $y_{B, \text{pH}} \approx 0.7$. The values of $j_{p,A}$ and $j_{p,B}$ both decrease with increasing pH.

A summary of the kinetic parameters of the benzoate system are given in Table 1, where they are compared with those obtained in acetate buffer[12]. Most noteworthy are the changes in the reaction orders in peak B. In the benzoate buffer, there is consistency between the reaction orders for peaks A and B, while this is not the case in the acetate system.

Much more information regarding the respective influences of acetate and benzoate on the anodic dissolution reactions can be obtained from experiments with mixtures. One series of anodic sweep voltammograms is seen in Fig. 3, for mixtures of pH 7 acetate and benzoate solutions, with a total buffer concentration [acetate] + [benzoate] = 1 M .

Table 1. Experimentally determined parameters describing the dissolution of Fe in benzoate and acetate[12] buffer solutions

Parameter	Benzoate	Acetate
$y_{A, [\text{buffer}]}$	≈ -1	-0.7 ± 0.2
$y_{B, [\text{buffer}]}$	≈ -1	≈ 0 (pH 7) > 0 ($5 \leq \text{pH} \leq 6$)
$\left(\frac{\partial \log j_{p,A}}{\partial \log [\text{buffer}]}\right)_{\text{pH}}$	≈ 0	$> 0^*$
$\left(\frac{\partial \log j_{p,B}}{\partial \log [\text{buffer}]}\right)_{\text{pH}}$	< 0	0 to 0.45^*
$\left(\frac{\partial E_{p,A}}{\partial \log [\text{buffer}]}\right)_{\text{pH}}$	> 0	small*
$\left(\frac{\partial E_{p,B}}{\partial \log [\text{buffer}]}\right)_{\text{pH}}$	> 0	small, $< 0^*$
$y_{A, \text{pH}}$	≈ 2	1.4 ± 0.2
$y_{B, \text{pH}}$	≈ 0.7	$< 0^*$
$\left(\frac{\partial \log j_{p,A}}{\partial \text{pH}}\right)_{[\text{buffer}]}$	< 0	-0.39 ± 0.1 (1 M) -0.22 ± 0.1 (0.1 M)
$\left(\frac{\partial \log j_{p,B}}{\partial \text{pH}}\right)_{[\text{buffer}]}$	< 0	-0.37 ± 0.1 (1 M) -0.15 ± 0.1 (0.1 M)
$\left(\frac{\partial E_{p,A}}{\partial \text{pH}}\right)_{[\text{buffer}]}$	< 0	-76 (1 M) -106 (0.1 M)
$\left(\frac{\partial E_{p,B}}{\partial \text{pH}}\right)_{[\text{buffer}]}$	< 0	-140 (1 M), -207 (0.1 M)
Tafel slope peak A	50 to 80	40 to 50
Tafel slope peak B	110 to 130	90 to 200

*No linear relationship

Fig. 3. Anodic sweep voltammograms for Fe at 5 mV s^{-1} in mixtures of acetate and benzoate buffer at pH 7. The total buffer concentration $[\text{acetate}] + [\text{benzoate}] = 1 \text{ M}$.

Clearly, the current density is much lower for pure benzoate buffer than for pure acetate buffer, and the mixture results show that small amount of added benzoate have a large effect on the anodic behavior. This result suggests that the benzoate species interact more strongly with the Fe surface to decrease the rate of Fe dissolution. A complementary experiment, which further highlights these differences between the two electrolytes is shown in Fig. 4. Here, the $[\text{acetate}]$ is held constant at 1 M, and $[\text{benzoate}]$ is varied. A comparison between Figs 3 and 4 clearly indicates that $[\text{benzoate}]$ is the critical variable in determining the current densities. At a given $[\text{benzoate}]$, there is very little influence of $[\text{acetate}]$. This result is confirmed by the experiments shown in Fig. 5, where $[\text{benzoate}]$ is held constant at 0.5 M, and $[\text{acetate}]$ is varied. Excluding those experiments where the buffer capacity is insufficient, *ie* $[\text{acetate}] + [\text{benzoate}] \leq 0.7 \text{ M}$, the voltammograms are virtually indistinguishable. Thus, it may be concluded that the role of $[\text{acetate}]$ in mixed solutions is as a supporting electrolyte, which simply provides sufficient conductivity and buffer capacity, and otherwise plays a negligible role in the dissolution reactions.

The voltammetric curves for benzoate were modelled using the same reaction scheme as has been used in the preceding paper for the acetate system[12] by substituting HB or B^- for HA or A^- . The reaction sequence is shown schematically in Fig. 6, and the reactions are detailed explicitly in Fig. 7. The data was fitted in a semi-quantitative manner, by adjusting

Fig. 4. Anodic sweep voltammograms for Fe at 5 mV s^{-1} in mixtures of acetate and benzoate buffer at pH 7, for $[\text{acetate}] = 1 \text{ M}$ with various $[\text{benzoate}]$.

Fig. 5. Anodic sweep voltammograms for Fe at 5 mV s^{-1} in mixtures of acetate and benzoate buffer at pH 7 for $[\text{benzoate}] = 0.5 \text{ M}$ with various $[\text{acetate}]$.

the rate and equilibrium constants in the model to obtain a reasonable correspondence ($\pm 40\%$) with the peak currents for pHs between 6.8 and 7.2, and concentrations between 0.75 and 1.5 M, and to give appropriate values of the reaction orders in peaks A and B. Details of the fitting procedure are given in the preceding paper[12]. In addition, the numerical values of the rate and equilibrium constants obtained in the simulation of the acetate data were changed as little as possible, specifically, only parameters for reactions in which HB or B^- were involved were permitted to vary. The parameters obtained for the benzoate system are shown in Table 2, where they are compared with those previously used to semi-quantitatively simulate the acetate data. As discussed in the preceding paper[12], the individual values of the rate and equilibrium constants are not expected to have any quantitative meaning. Nevertheless, the model can be used to predict coverages, θ_i , and trends with respect to solution composition, as will be discussed below. Simulations of the data in Figs 1 and 2 are shown in Figs 8 and 9. It can be seen that there is reasonable agreement between the respective data sets, particularly with respect to the peak currents and reaction orders with respect to pH and $[\text{benzoate}]$. Two features of the experimental data are not particularly well represented by the simulation. The first is the absolute potential scale, and the second is the definition of the minimum between peaks A and B which is less distinct in the simulated data. For reasons discussed in the preceding paper[12], the model is expected to provide, at best, a semi-quantitative fit to the experimental data. In the present work, the anodic currents are at least an order of magnitude less than in the preceding work. This means that a significant fraction of the current is going towards oxide formation, while the model assumes only a steady state current corresponding to faradaic Fe dissolution. For this additional reason, a perfect fit to the data is not expected.

Figure 10 shows the coverages of the various species in the model for pH 7, (a) $[\text{acetate}] = 1 \text{ M}$ and (b) $[\text{benzoate}] = 1 \text{ M}$. Comparison of Fig. 10a and b indicates that the coverage θ_2 persists to higher potentials in the case of benzoate, originating from a decrease in K_5 (see Table 2). This results in lower currents in peak A in the case of benzoate, and a shift of the Tafel region of peak A to more anodic

Fig. 6. A schematic of the equilibria considered in the proposed reaction mechanism. The species considered are enclosed in boxes, and are assigned a surface coverage θ_i , $i = 1$ to 8. The arrows connecting the boxes refer to the equilibria which are assumed to exist between the various species. The large end of the arrow indicates the direction in which the reaction would be shifted for a large value of the equilibrium constant, K_i , $i = 1-6, 9-15$. The heavy line indicates the reaction path followed for the results of the fit to experimental data for benzoate solutions.

potentials. Both these trends are visible in the experimental data (see for instance Fig. 3). A physical interpretation of this modification is that adsorption of HB on Fe is so strong that it inhibits the oxidation reaction, K_2 . The other major difference between Fig. 10a and b is an increase in the coverage of θ_6 relative to that of θ_5 , in the case of benzoate. This is a direct result of the increase in the value of K_{10} , see Table 2. This modification to the parameters results in a negative value of $y_{B, [\text{benzoate}]}$, and it can be physically interpreted as a blockage of the surface by the species $[\text{Fe}(\text{OH})(\text{B})]_{\text{ad}}$ which does not react to yield anodic current. Finally, as seen in Table 2, the value of the rate constant k_{17} was set to zero in the case of benzoate. This reaction corresponded to an acceleration of the dissolution in the region of peak B by the anion in solution. Since $y_{B, [\text{benzoate}]}$ is less than zero, this reaction is not required in the overall scheme for benzoate. While there is appreciable coverage of all species (except θ_8) in the case of acetate at various pH and [acetate] (although the coverage of θ_4 is negligible

in Fig. 10a, it is appreciable at lower [acetate] and pH, see preceding paper [12]) the coverage of θ_1 , θ_4 , and θ_8 are all negligible in the case of benzoate. This means that while the reaction flows through all of the possible paths given in Fig. 6 for the case of acetate, in the case of benzoate the reaction follows only the route shown by the heavy lines in the figure, indicating that a simpler reaction scheme is sufficient to describe the data in the case of benzoate. Nevertheless, the full reaction scheme is general enough to fully describe the mechanism of Fe dissolution in both acetate and benzoate buffer solutions.

By the same procedure as used in the preceding paper in acetate buffer [12], galvanostatic cathodic reduction has been used to characterize the oxide film present on the Fe surface during the course of anodic sweep voltammetry in benzoate buffer. As was the case in pH 7, [acetate] = 1 M, the arrest structure could be divided into two regions. The first arrest region, referred to as Q_1 , was concluded to result from an Fe_2O_3 (or Fe^{III} -containing) oxide, while the

Fig. 7. Equilibrium reactions (equations 1–6, 9–15) and rate determining steps (rds, equations 7, 8, 16–18) considered in the proposed mechanism, shown schematically in Fig. 6. Fe, represents an iron atom in the metallic substrate. The equilibrium constant K_i is given by k_i/k_{-i} for $i = 1-6, 9-15$.

Fig. 8. Simulated anodic sweep voltammograms for Fe in pH 7 solution with [benzoate] = 0.75, 1.0 and 1.5 M. The constants shown in Table 2 were used in the simulation. The experimental data are shown in Fig. 1.

Fig. 9. Simulated anodic sweep voltammograms for Fe in pH 7.2, 7.0, and 6.8 solution with [benzoate] = 1 M. The constants shown in Table 2 were used in the simulation. The experimental data are shown in Fig. 2.

Fig. 10. The surface coverages, θ_i , $i = 1$ to 8, calculated from the model for (a) pH 7, [acetate] = 1 M, and (b) pH 7, [benzoate] = 1 M. θ_4 and θ_8 are negligible in both simulations, and θ_1 is negligible for benzoate. The constants shown in Table 2 were used in the simulations.

Table 2. Values of the rate and equilibrium constants used in the simulation using the model illustrated in Fig. 6 and 7

Independent equilibrium constants			Derived equilibrium constants			Rate constants for rds		
	Acetate	Benzoate		Acetate	Benzoate		Acetate	Benzoate
K_1	1500	1500	K_2	1.17×10^6	1.4×10^{10}	k_7	2.9×10^{-5}	2.9×10^{-5}
K_4	700	700	K_3	778	9.33×10^6	k_8	5×10^{-4}	5×10^{-4}
K_5	0.6	5×10^{-5}	K_{11}	105	3×10^4	k_{16}	1×10^{-10}	2.5×10^{-7}
K_6	1.5	1.5	K_{13}	52.5	1.5×10^4	k_{17}	6×10^{-7}	0
K_9	20	20	K_{14}	0.286	1×10^{-3}	k_{18}	1×10^{-7}	1×10^{-7}
K_{10}	70	2×10^4						
K_{12}	2	2						
K_{15}	1×10^{-4}	1×10^{-4}

Fig. 11. (a) Experimental anodic voltammogram for Fe at 5 mV s^{-1} in pH 7 solution with $[\text{benzoate}] = 1 \text{ M}$. (b) The cathodic charges corresponding to the two arrests in the galvanostatic cathodic reduction profile. The potential was swept to a given value at 5 mV s^{-1} , and then galvanostatic reduction at $20 \mu\text{A cm}^{-2}$ was started. The charge Q_1 refers to reduction of a Fe^{III} -containing oxide, while Q_2 refers to reduction of a Fe^{II} -containing oxide.

second arrest region, with cathodic charge Q_2 , corresponded to reduction of Fe_3O_4 or some other Fe^{II} -containing oxide. Figure 11a shows the anodic sweep result for pH 7, $[\text{benzoate}] = 1 \text{ M}$ at 5 mV s^{-1} . Figure 11b shows the cathodic charges in the first and second arrest regions as a function of the potential at which the anodic sweep was interrupted and the galvanostatic cathodic charging at $20 \mu\text{A cm}^{-2}$ begun. It can be seen that as the anodic current rises in the Tafel region of peak A, an arrest corresponding to an Fe^{II} -containing oxide (Q_2) begins to appear which grows in magnitude through peak A. After the current has decreased in the transition to passivity, the arrest corresponding to the Fe^{III} -containing oxide (Q_1) appears, and increases steadily with potential. Unfortunately, it is difficult to quantify the exact coverages corresponding to the values of cathodic charge because the reduction efficiencies for the various oxides in benzoate solution are not known. Nevertheless, it is apparent that there is a strong correlation between Q_2 and the coverage θ_3 (or θ_6) and between Q_1 and the coverage θ_7 . As in the preceding work[12], these results provide direct, *in situ* electrochemical evidence for the coverage of the surface by a Fe^{II} -containing oxide during the active dissolution in peaks A and B, and thus provide support for the proposed model. Again, as in the preceding work, the striking difference between the model calculation and the experimental results is that Q_2 does not disappear as Q_1 increases, in other words, experimentally, the Fe^{III} -containing oxide is formed on top of the Fe^{II} -containing oxide, rather than

replacing the lower oxide. This illustrates an important limitation of the model; since the maximum coverage is assumed to be unity, the model cannot account for a true phase oxide. It is not expected, therefore, that the model will satisfactorily describe the transition between peak B and passivation.

SUMMARY

Potential sweep experiments, coupled with galvanostatic cathodic reduction, have been used to study the kinetics of the dissolution and passivation of Fe in benzoate solutions, and the results have been compared with those obtained previously in acetate solutions. The results suggest that the significant suppression of the anodic current in benzoate solution arises from a strong interaction between benzoate solution species and the Fe surface. Negative reaction orders with respect to total benzoate concentration, and positive reaction orders with respect to pH, were observed for the Tafel regions of two anodic peaks in the active dissolution region of the voltammograms. Experiments in acetate/benzoate mixtures established that the total benzoate concentration is the critical variable in determining the anodic sweep voltammogram, and that the role of acetate in the mixture is as a supporting electrolyte which simply provides sufficient conductivity and buffer capacity, but otherwise plays a negligible role in the dissolution reactions. The same mechanism which had been used to fit the anodic data for the acetate buffer system was successfully used to provide a semi-quantitative fit to the data for benzoate solutions. A better fit to the data would likely result if rigorous, steady-state conditions had been reached. The variation in the kinetic parameters could be explained by simple, physical interpretations involving a stronger interaction between the benzoate solution species and the Fe surface than with the corresponding acetate species.

REFERENCES

1. M. J. Pryor and M. Cohen, *J. electrochem. Soc.* **100**, 203 (1953).
2. D. M. Brasher and A. D. Mercer, *Br. Corros. J.* **3**, 120 (1968).
3. R. Natarajan and A. D. Purohit, *Indian J. Technol.* **8**, 98 (1970).
4. D. E. Davies and Q. J. M. Slaiman, *Corros. Sci.* **11**, 671 (1971).
5. Q. J. M. Slaiman and D. E. Davies, *Corros. Sci.* **11**, 683 (1971).
6. J. C. Wood and N.-G. Vannerberg, *Corros. Sci.* **18**, 315 (1978).
7. G. N. Mehta and J. P. Sastry, *Trans. Soc. Adv. Electrochem. Soc. Technol.* **13**, 31 (1978).
8. I. L. Rozenfel'd, Yu. I. Kuznetsov, I. Ya. Kerveleva, N. N. Balashova and N. A. Solomko, *Zashch. Met.* **14**, 343 (1978).
9. S. Sanyal and B. Sanyal, *Indian J. Technol.* **24**, 663 (1986).
10. S. Turgoose, *Proc. 6th Eur. Symp. Corros. Inhib.* Ferrara, Italy, 1985, p. 1041.
11. Yu. I. Kuznetsov and N. N. Andreev, *Zashch. Metal.* **23**, 495 (1987).
12. K. Takahashi, J. A. Bardwell, B. MacDougall and M. J. Graham, *Electrochim. Acta* **37**, 477 (1992).