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Autocatalytic dissolution of Pb in HNO_3

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The reaction between Pb and HNO_3 has been investigated using the thermometric technique. Weight-loss measurements on the reaction were also obtained. As the HNO_3 concentration is increased from 5×10^{-2} to 4 mol l^{-1}, the corrosion rate increases. This is shown thermometrically by a substantial increase in the maximum temperature attained, T_m, as well as a decrease in the time, *t*, required for reaching T_m. Dissolution of Pb in HNO_3 is proposed to take place according to an autocatalytic mechanism. Passivation sets were detected in solutions $\geqslant 11$ mol l^{-1} HNO_3. A parallel indication between the thermometric technique and weight-loss measurements was obtained. The rate-determining step of the autocatalytic process involves HNO_2 in dissolution of Pb in HNO_3. This is supported by the results of addition of hydrazine to the solution. This additive raises the maximum measured temperature, without affecting the corresponding time necessary to reach it. The effect of addition of $NaNO_2$, $NaNO_3$, NaCl, Na_2SO_4, NaH_2PO_4 and $NaClO_4$ on the reaction number, RN, of Pb in 4 mol l^{-1} HNO_3 was examined. Only $NaNO_2$ accelerates the dissolution reaction while the other salts show as inhibition effect. It was found that these additives inhibit dissolution due to the displacement of some cathodic depolarizing components, as NO_2, from the active sites on the metal surface. The effect of addition of HCl, H_2SO_4, $HClO_4$ and H_3PO_4 on the reaction number, RN, of Pb in 4 mol l^{-1} HNO_3 was also investigated. The observed acceleration and retardation of the dissolution of Pb was found to be dependent on both the concentration and nature of anions of the extra acids added.

1. Introduction

The rate of dissolution of metals in HNO_3 is much higher than those in other acids of comparable concentration. This is a result of the occurrence of an autocatalytic process [1] involving the production of HNO_2. According to Smolyaninov [2], however, it was found that the rate of dissolution of lead in HNO_3 solution increases with increasing acid concentration up to a certain point, whereafter the reaction is inhibited by the formation of an adherent film of $Pb(NO_3)_2$ which is insoluble in HNO_3 and, moreover, is changed to a more protective film of PbO_2 at higher concentration of the acid [3].

The present work reports on the corrosion and passivation behaviour of Pb in HNO_3. The influence of various additives and the implications of the results on the dissolution mechanism is discussed. Since the reaction between Pb and HNO_3 involves a large decrease in free energy, it is of interest to study the behaviour of this system by the thermometric technique [4]. This technique allows certain features of corrosion and corrosion inhibition to be easily examined. Some of the reactions were also studied using the weight-loss technique.

2. Experimental procedure

The reaction vessel and the procedure for applying the thermometric method for following the dissolution of metals in different media have been described previously [5]. The lead used was supplied by the Egyptian Company for Metals Trading (SIGAL). Lead test pieces in the form of sheets having a geometrical surface area of 12.5 cm^2 were used. Before being used, these were abraded successively with 0, 00, and 000 emery paper, then degreased with acetone and dried between filter papers. This treatment was carried out immediately before making the test. Each experiment was carried out with 15 ml of the acid solution and with a fresh test piece. The temperature was measured to ± 0.1 °C. All chemicals used were of AR quality and were employed without further purification. The molar concentration of each solution was checked against standard NaOH. The initial temperature of all the experiments was 20 ± 0.1 °C.

The rate of reaction is characterized by the reaction number, RN, in °C min^{-1} which is defined as [4, 5]

$$RN = (T_m - T_i)/t \qquad (1)$$

where T_m and T_i are the maximum and initial temper-

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ature, respectively, and t is the time in minutes taken to attain T_m. Weight-loss experiments were performed under similar conditions to the thermometric measurements for 90 min. Duplicate experiments were carried out and weight-losses were averaged.

3. Results and discussion

The curves in Fig. 1 represent the variation of temperature with time for the system when Pb test pieces of 12.5 cm^2 were made to react with 15 ml of HNO_3 of different concentrations. The curves in this figure are separated into two groups exhibiting two different behaviours. Group a includes the curves obtained with HNO_3 from 5×10^{-2} to 4 mol l^{-1}, while group b from 4.5 to 13 mol l^{-1} HNO_3. The direction of the change of the RN and weight-loss values with the HNO_3 concentration is represented in Fig. 2. Up to a concentration of 0.1 mol l^{-1} HNO_3 no temperature change was recorded, Fig. 2a, while slight dissolution was observed, Fig. 2b. In solutions of 0.5 to 2 mol l^{-1} HNO_3, a slight temperature increase with time was

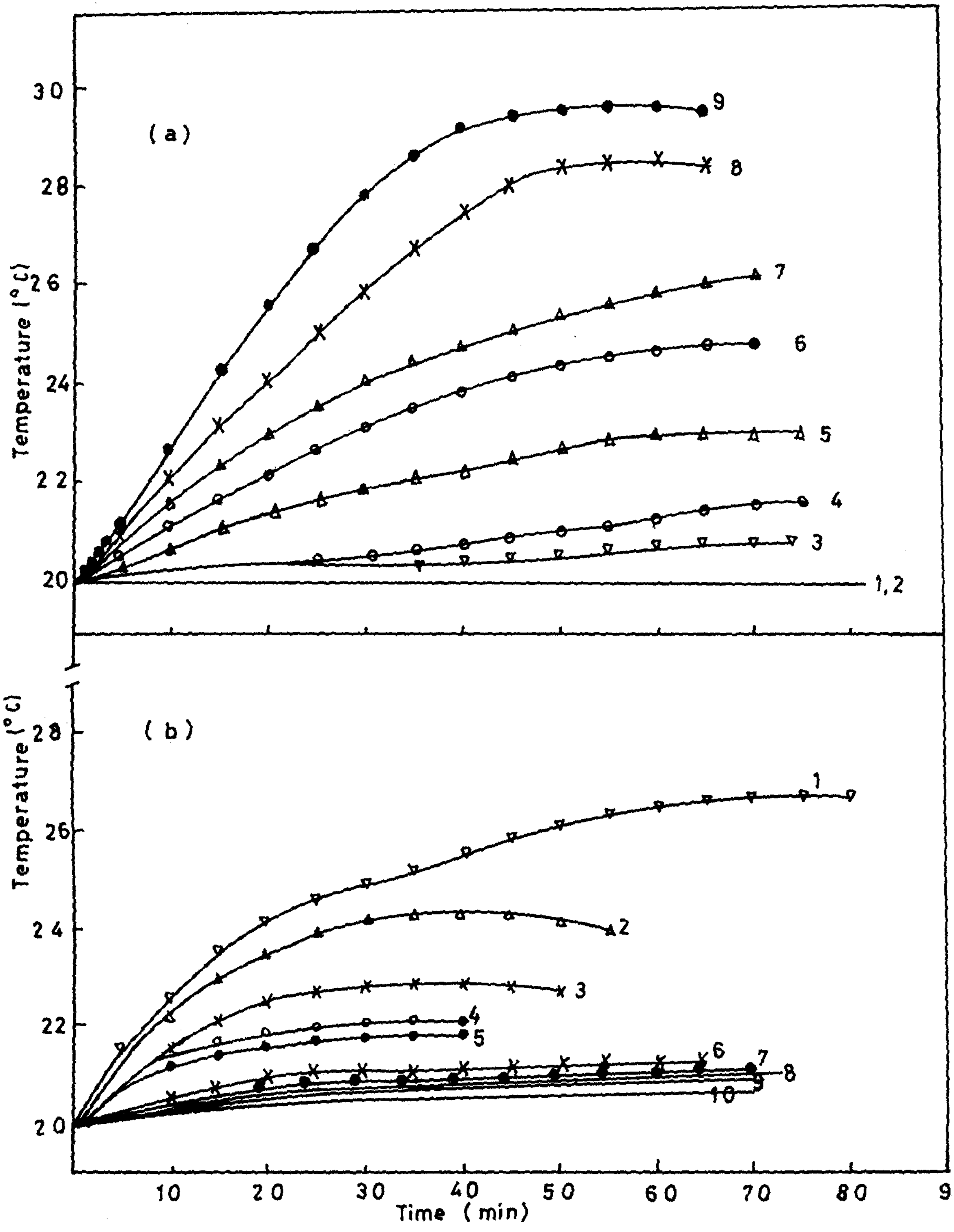

Figure 1 Temperature–time curves for the reaction of Pb with solutions of HNO_3 of concentrations (a) from 5×10^{-2} to 4 mol l^{-1}; (1, 5×10^{-2}; 2, 1×10^{-1}; 3, 5×10^{-1}; 4, 1.0; 5, 2.0; 6, 2.5; 7, 3.0; 8, 3.5; 9, 4.0); (b) from 4.5 to 13 mol l^{-1} (1, 4.5; 2, 5.0; 3, 6.0; 4, 7.0; 5, 8.0; 6, 9.0; 7, 10; 8, 11; 9, 12; 10, 13).

observed until the maximum temperature, T_m, is attained. As the acid concentration is further increased, the maximum temperature during the course of the reaction increases while the time, t, required to reach T_m decreases. This behaviour indicates that the dissolution rate of Pb increases with increasing the concentration of HNO_3. As seen from Fig. 2a and b, the RN and weight-loss increase with acid concentration, and reach a maximum at ~ 4 mol l^{-1} HNO_3. This value is in fair agreement with that given by Sayed *et al.* [6], who reported that the highest rate of the metal dissolution was 3.1 mol l^{-1} HNO_3. Increasing the concentration of the acid > 4 mol l^{-1} HNO_3, Fig. 1b, causes a retardation of the dissolution process. This can be seen from the decrease of the maximum temperature value and the increase of the time taken to attain that maximum.

From the plot of the RN as a function of the molar

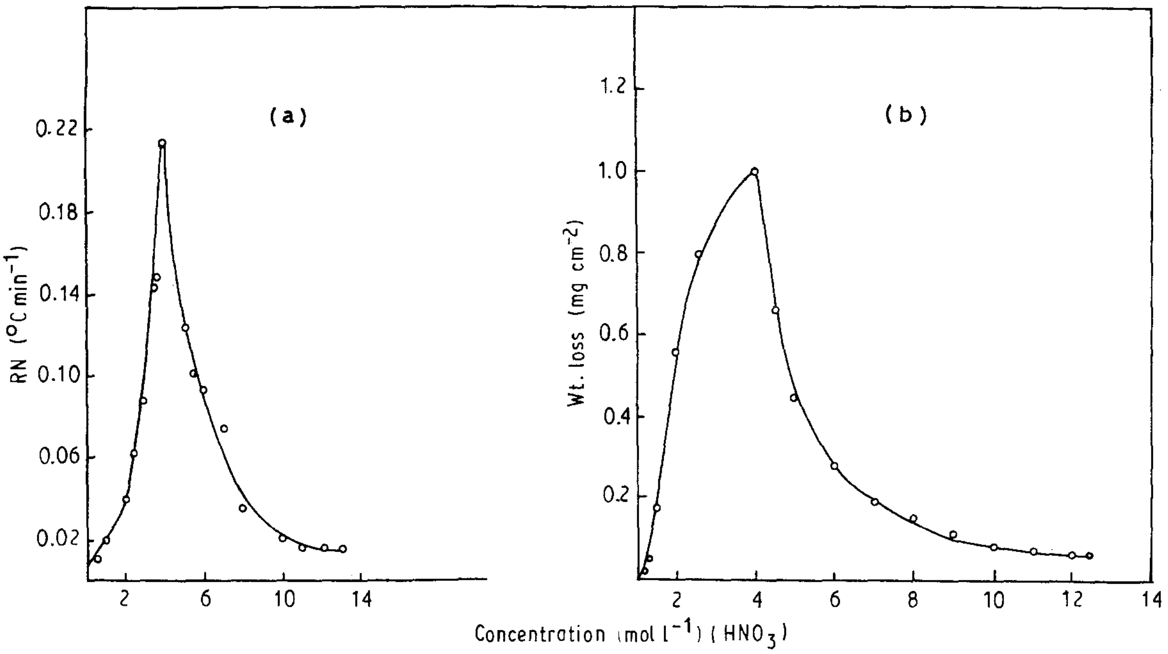

Figure 2 Variation of (a) the RN and (b) Pb weight-loss with various concentrations of HNO_3.

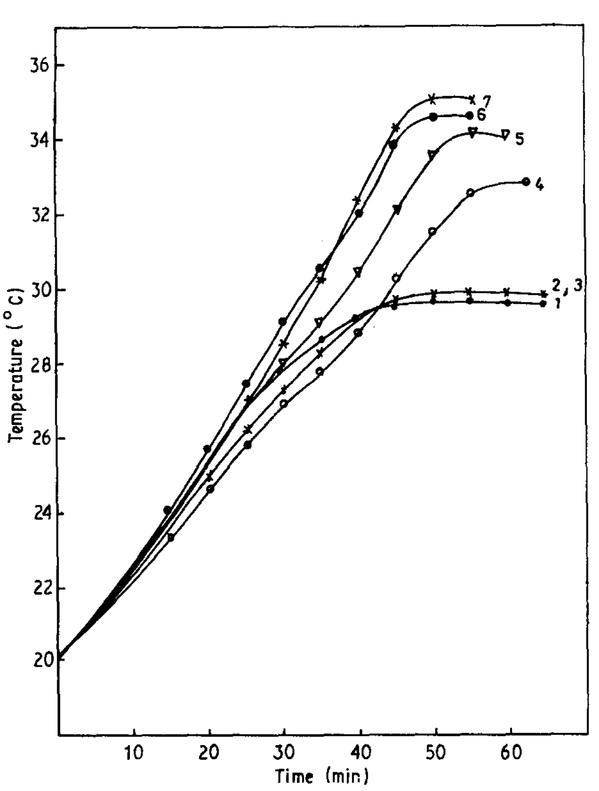

Figure 3 Effect of the addition of hydrazine (mol l^{-1}) (1, 0; 2, 1×10^{-3}; 3, 1×10^{-2}, 4, 5×10^{-2}; 5, 7×10^{-2}; 6, 9×10^{-2}; 7, 2.5 $\times10^{-1}$) on the thermometric behaviour of Pb in 4 mol l^{-1} HNO_3.

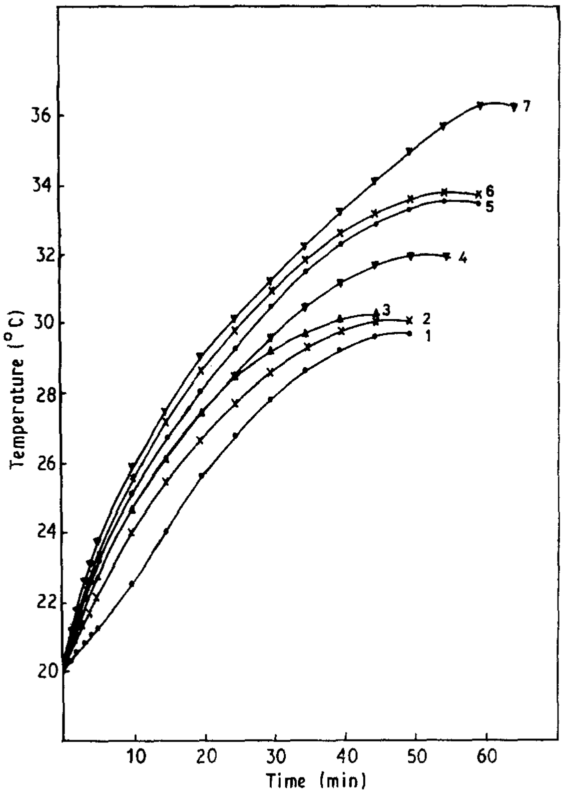

Figure 4 Effect of the addition of $NaNO_2$ (mol l^{-1}) (1, 0; 2, 1.0 $\times10^{-3}$; 3, 1.0×10^{-2}; 4, 6.0×10^{-2}; 5, 8.0×10^{-2}; 6, 1.0×10^{-1}; 7, 2.0 $\times10^{-1}$) on the thermometric behaviour of Pb in 4 mol l^{-1} HNO_3.

concentration of HNO_3 solution (Fig. 2a) it is interesting to note that the rate of dissolution of Pb increases slowly in concentration from 0.5 to 2 $mol\,l^{-1}$ HNO_3 and then jumps abruptly to a maximum at an acid concentration of 4 $mol\,l^{-1}$. Thereafter, the RN drops to an almost constant lower value in concentration from 11 to 13 $mol\,l^{-1}$ HNO_3. The fact that the curves of RN and weight-loss (Fig. 2a and b) exhibit a maximum, indicates that the dissolution of Pb in HNO_3 is controlled by two opposing reactions. The first, favouring corrosion and its rate increases with the increase in the acid concentration. The second reaction favours passivity and is caused by the increased oxidizing properties of the medium as the acid concentration is raised. This leads to an enhancement of the rate of formation and repair of a passivating film of PbO_2 on the metal surface [6]. It is known that $Pb(NO_3)_2$ is changed to PbO_2 at higher concentration of HNO_3 [7]. The stability and thickness of this passive film formed in such solutions increases with acid concentration more rapidly than the rate of its dissolution [6]. The overall rate of lead dissolution and consequently the RN and weight-loss values therefore steadily decrease, Fig. 2.

In solutions $\geqslant 11$ $mol\,l^{-1}$ HNO_3, the dissolution reaction practically ceases. In these concentrated solutions, the passivation takes place very shortly after immersion of the lead specimen in the acid solution. The curves given in Fig. 1b show, however, that over the entire span of the experiments, a temperature rise of 0.6 to 1.1 °C is always measured. This slight temperature rise is due to the heat evolved from the reaction leading to the healing of imperfections and strengthening of the passivating film. Such imperfections would arise from acid attack and/or reductive dissolution [8].

The dissolution of lead in HNO_3 solutions along the rising parts of the curve, Fig. 1, may take place according to [9]

$$3Pb + 8HNO_3 = 3Pb(NO_3)_2 + 2NO + 4H_2O \qquad (2)$$

Due to the high hydrogen overvoltage of lead [10], the partial cathodic reaction corresponding to such dissolution rate, would be related to the reduction of HNO_3 rather than the evolution of hydrogen. The cathodic reduction of nitric acid by atomic H leads to a considerable decrease of free energy which depolarizes the overall dissolution process. This is preceded by the primary displacement of H^+ ions from the solution according to

$$H^+ + e^- = H \qquad (3)$$

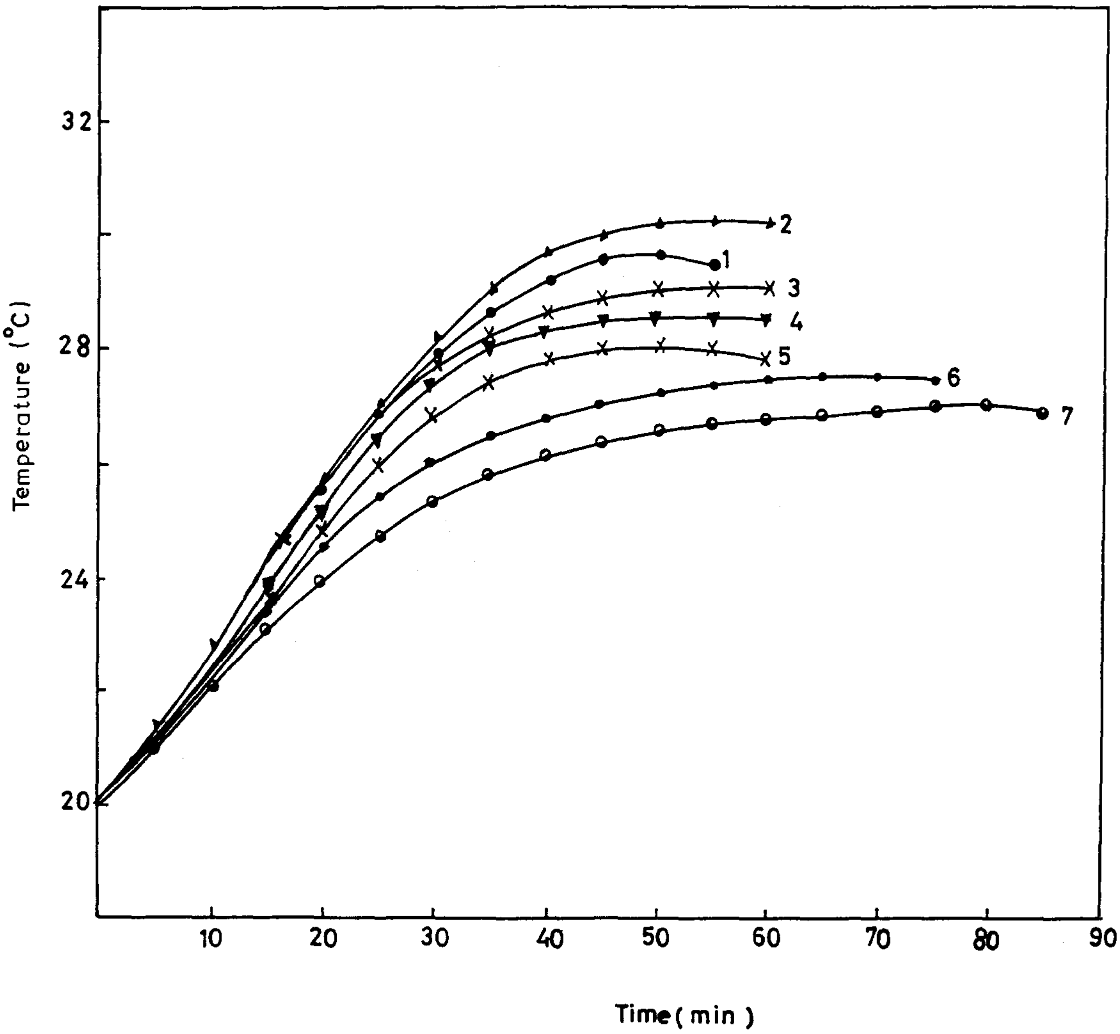

Figure 5 Effect of the addition of $NaNO_3$ ($mol\,l^{-1}$) (1, 0; 2, 5.0×10^{-2}; 3, 1.0×10^{-1}; 4, 2.5×10^{-1}; 5, 5.0×10^{-1}; 6, 1.6; 7, 2.0) on the thermometric behaviour of Pb in 4 $mol\,l^{-1}$ HNO_3.

$$H + HNO_3 = H_2O + NO_2 \quad (4)$$

The NO_2 molecules formed would then be adsorbed on the metal surface, taken up an electron, and undergo the following reactions:

$$NO_2 + e^- = NO_2^- \quad (5)$$

$$H^+ + NO_2^- = HNO_2 \quad (6)$$

$$HNO_2 + HNO_3 = 2NO_2 + H_2O \quad (7)$$

This means that, in strong acid solutions, each molecule of NO_2 produces two NO_2 molecules. The action therefore becomes self-stimulating and rapid, providing that chain propagation mechanism takes place. This would explain the increase in the slope of the raising part of the curve which relates RN to acid concentration (Fig. 2a). Similar findings were proposed to explain the dissolution of Fe [8], Cu [11, 12] and Sn [13] in HNO_3. The retardation of lead dissolution in relatively concentrated HNO_3 solutions is attributed to the formation of a covering layer of insoluble PbO_2 and/or a protective film of lead nitrate [6, 9] on the surface of the metal which prevents further dissolution.

The formation of HNO_2 as the rate-determining step in the mechanism of Pb dissolution in HNO_3 is verified in the present work by the addition of some additives on the thermometric behaviour of Pb in $4\,\text{mol}\,l^{-1}$ HNO_3. This acid concentration represents the range where Pb is assumed to dissolve in HNO_3 according to the autocatalytic mechanism (Equations 2 to 7).

The HNO_2 is involved in the autocatalytic cycle of metal dissolution in HNO_3. This was shown from the effect produced by hydrazine on the rate of the reaction. The additive destroys HNO_2 (Equation 6) according to a reaction of the type [13, 14]:

$$2H_2N\text{-}NH_2 + 2HNO_2 = 3N_2 + H_2 + 4H_2O \quad (8)$$

The curves of Fig. 3 represent the effect of increasing the concentration of hydrazine on the thermometric curves of Pb in $4\,\text{mol}\,l^{-1}$ HNO_3. The interesting feature of these curves is that at lower hydrazine concentrations (10^{-3} and $10^{-2}\,\text{mol}\,l^{-1}$), it has practically no effect on the rate of metal dissolution. This behaviour can be explained on the basis that both T_m and the time taken to reach T_m always remains the same as in additive-free solutions. As the concentration of the hydrazine is further increased, (5×10^{-2} to $1 \times 10^{-1}\,\text{mol}\,l^{-1}$), the curves obtained exhibit an increase in temperature above that measured in additive-free solutions, consequently increasing the

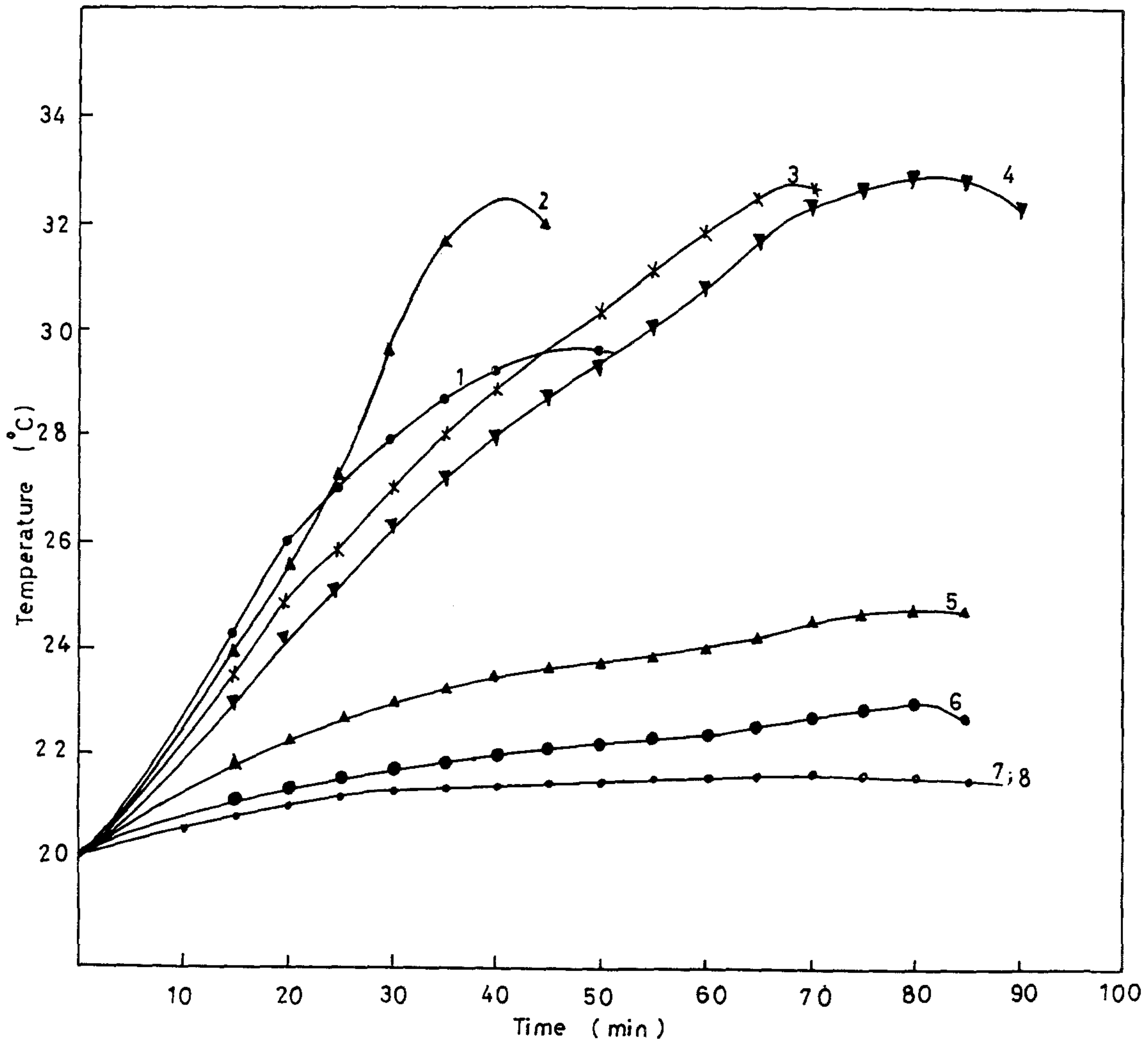

Figure 6 Effect of the addition of NaCl ($\text{mol}\,l^{-1}$) (1, 0; 2, 1×10^{-2}; 3, 6×10^{-2}; 4, 8×10^{-2}; 5, 1×10^{-1}; 6, 1.5×10^{-1}; 7, 2×10^{-1}; 8, 2.5×10^{-1}) on the thermometric behaviour of Pb in $4\,\text{mol}\,l^{-1}$ HNO_3.

dissolution rate of Pb. Under the conditions of our experiments, where the volume and strength of HNO_3, and also the area of corroding metal are kept the same, the rate of production of HNO_2 should remain constant [11]. The rise in temperature therefore depends on the concentration of hydrazine. The rise in temperature when further concentration of hydrazine was added can be explained on the basis of two reasons [6]. Firstly, the reaction between hydrazine and HNO_2 (Equation 6) is associated with rise in temperature, this rise in temperature will be increased with further concentration of one of reactants (hydrazine, for the acid concentration to remain constant). This heat is added to the heat of dissolution of the metal without additive. Secondly, hydrazine may be adsorbed on the metal surface producing a protective layer, which prevents the formation of PbO_2 and permits the reaction of lead with HNO_3 to take place, causing the promotion of corrosion and consequently a rise in the maximum temperature. Similar conclusions were found to describe the dissolution of Pb under similar conditions using the urea as additive by Sayed *et al.* [6].

The effect of addition of a number of inorganic compounds such as $NaNO_2$, $NaNO_3$, NaCl, Na_2SO_4, $NaClO_4$ and NaH_2PO_4, on the thermometric behaviour of Pb in 4 $mol\,l^{-1}$ HNO_3 was investigated. These effects are described as follows. According to Fig. 4, as the concentration of NO_2^- ion increases, the maximum temperature, T_m, increases and the time, t, taken to attain that T_m decreases. Therefore, the addition of NO_2^- ion accelerates the dissolution reaction of Pb. This suggests that NO_2^- or one of its direct reduction products is involved in the rate-determining step, so that the addition of NO_2^- accelerates the overall process by the production of HNO_2. Although NO_2^- accelerates corrosion in acid media, it is widely used as a corrosion inhibitor in alkaline and neutral solutions [15].

The effect of the addition of NO_3^- ion on the course of the reaction is shown in Fig. 5. It is noted that the addition of a dilute concentration of NO_3^- ions leads to a decrease in the time, t, taken to reach T_m while the latter remains more or less unchanged as in NO_3^--free solution. While the concentration of NO_3^- ion increases the dissolution rate of Pb decreases, since T_m decreases and the time, t, taken to attain that T_m increases. This clearly indicates that the NO_3^- ion is not directly involved in the rate-determining step. The fact that the increase of the NO_3^- ion inhibits the dissolution of Pb may be attributed to displacement of some cathodic depolarizing component such as NO_2, from the active sites on the metal surface. On the other hand, this inhibition effect may be also due to the

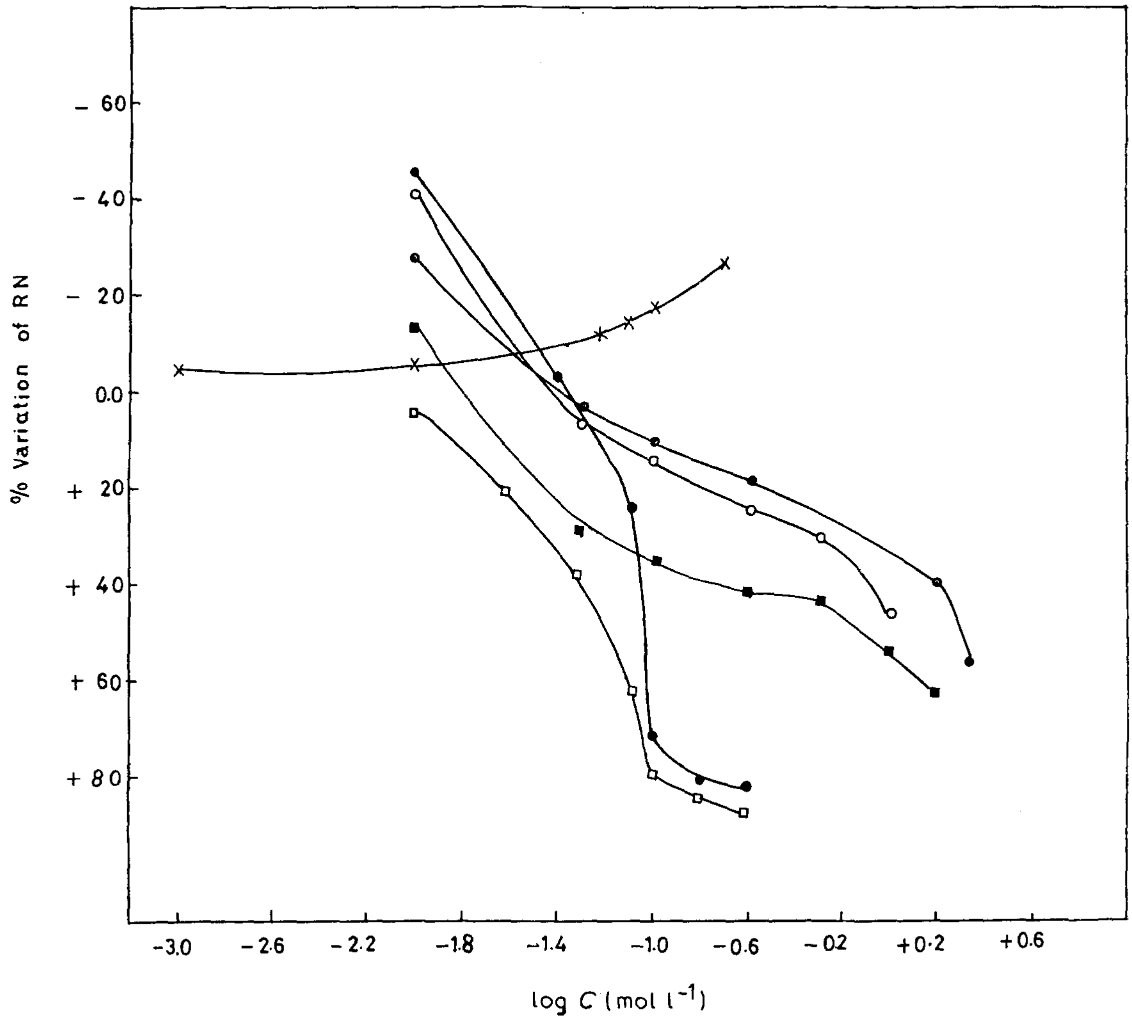

Figure 7 The percentage variation of the RN with the concentration of various inorganic salt additives of Pb in 4 $mol\,l^{-1}$ HNO_3. (× $NaNO_2$, ⊙ $NaNO_3$, ○ NaH_2PO_4, ■ $NaClO_4$, ● NaCl, □ Na_2SO_4).

reduction of NO_3^- ion on the surface of the metal according to a reaction of the type [16]

$$NO_3^- + 10H^+ + 8e^- = NH_4^+ + 3H_2O \quad (9)$$

This type of reaction clearly demonstrates that the excess of NO_3^- ion, consume H^+ ions at the electrode surface level in the ratio of $10H^+$ to each NO_3^- ion. This will hinder the chain propagation of the autocatalytic mechanism leading to the observed retardation [13].

The Cl^- ion, which normally acts as a pitting corrosion agent, functions in the present case as a retardant. In the presence of low Cl^- ion concentrations (Fig. 6), the curves obtained exhibit an increase in maximum temperature, T_m, above that measured in Cl^--free solution while the time taken to reach, T_m, decreases, the rising parts of the temperature–time curves are parallel to that obtained in pure HNO_3. Since, according to Equation 1, the slope of this part of the temperature–time curves is a measure of the actual rate of anodic dissolution [8]. The observed influence of Cl^- ion is, therefore, due to its interaction with the partial cathodic process [14]. The results can be tentatively explained on the basis of the occurrence of competitive adsorption on the metal surface. It is assumed that Cl^- ions displace NO_2 from active sites. NO_2 acts as cathodic depolarizer whilst Cl^- does not, hence retardation of the dissolution takes place. On the other hand, as the concentration of Cl^- ion increases, the maximum temperature, T_m, decreases and the time, t, taken to reach that T_m increases. Furthermore, the rising parts of the temperature curve is no longer parallel to that obtained for Cl^--free solutions. Cl^- ions here would influence also on the anodic reaction of Pb in HNO_3 [17, 18]. Similar behaviour was obtained by the thermometric curves in the presence of SO_4^{2-}, ClO_4^- and $H_2PO_4^-$. The results are best represented by Fig. 7, which gives the percentage variation in reaction number (% RN) according to the known formula

$$\frac{(RN)_{add} - (RN)_{free}}{(RN)_{free}} \times 100, \text{ of Pb in } 4\,mol\,l^{-1}\,HNO_3$$

The (% RN) values are plotted as a function of the logarithm of the molar concentration of the salt.

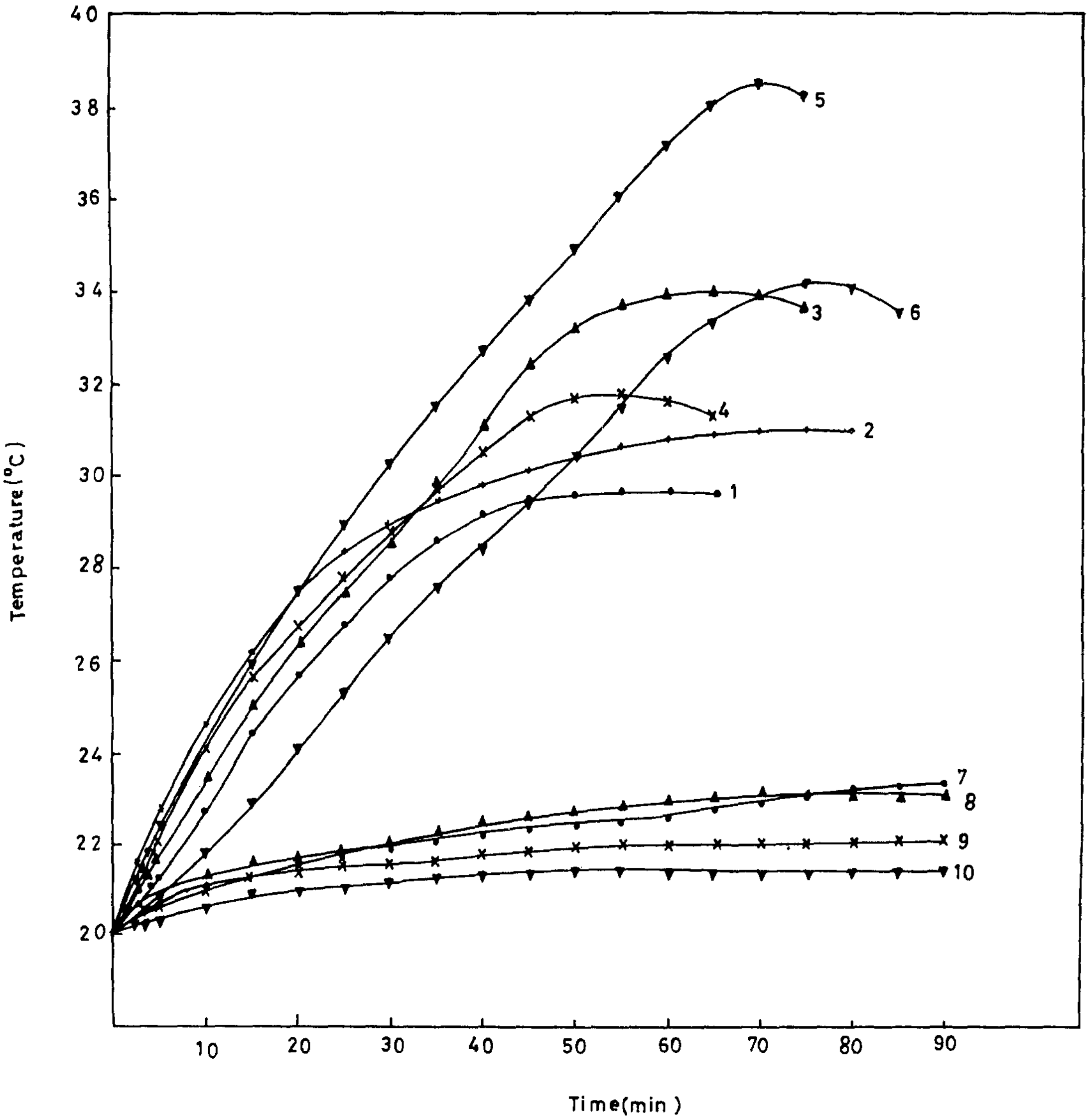

Figure 8 Effect of the addition of HCl ($mol\,l^{-1}$) (1, 0; 2, 1.0×10^{-2}; 3, 5.0×10^{-2}, 4, 6.0×10^{-2}; 5, 8×10^{-2}; 6, 9.0×10^{-2}; 7, 1.2×10^{-1}; 8, 1.5×10^{-1}; 9, 5.0×10^{-1}; 10, 1.00) on the thermometric behaviour of Pb in $4\,mol\,l^{-1}$ HNO_3.

The dissolution of Pb in HNO_3 is inhibited by the added anions according to the order SO_4^{2-} (strong) $> Cl^- > ClO_4^- > H_2PO_4^- > NO_3^-$ (weak). Both the tendency of adsorption of these anions and the solubility [9] of the corresponding Pb salts are expected to determine the extent of their inhibition of Pb dissolution.

To gain insight into the mechanism of dissolution of Pb in HNO_3, the effect of a number of acids on the dissolution of Pb in 4 $mol\,l^{-1}$ HNO_3 was also investigated. In Fig. 8, curves represent the thermometric behaviour of Pb in 4 $mol\,l^{-1}$ HNO_3 to which increasing concentration of HCl were added. Similar curves were obtained with additions of H_2SO_4, $HClO_4$ and H_3PO_4. The addition of extra acids was found to influence the percentage variation of RN (Fig. 9) in one of two ways. The addition of small amounts of extra acids (to the base solution of 4 $mol\,l^{-1}$ HNO_3) brings about a decrease in % RN (acceleration) caused mainly through a diminution in the time, t, necessary to reach T_m, while the value of T_m, is greater than that of free HNO_3. As the concentration of extra acid in solution is increased, retardation is augmented and higher % RN (inhibition) values are obtained. The increase in % RN is brought about through a decrease in T_m and an increase in t. The retardation effect will increase with further addition of extra acid.

The four acids thus act either as accelerators or as inhibitors for the dissolution of Pb in HNO_3, depending upon their concentration in solution. On the addition of a small concentration of the extra acid, acceleration of dissolution of Pb is obtained. This is attributed to the increase in the acidity of the solution [6]. The addition of more concentrated solution of extra acids, however, causes retardation of the dissolution of Pb. This may be explained on the basis that the increase in acidity of the solution favours the formation of PbO_2 which is more resistant to dissolution [6]. On the other hand, the adsorption of the anion of the extra acid on the surface of the Pb displaces one of the reaction products of the NO_3^- ions, involved in the autocatalytic cycles, and also leads to the retardation of the dissolution reaction [11]. Since the energies of adsorption of the various anions on the surface of the metal depend on the nature of the specific anion [11, 19]; also since the retardation effect varies from one acid to another; it can, therefore, be concluded that the extent of retardation presumably depends on the nature of the anion of the extra acid added. From the effect of various extra acid additions on the % RN of Pb in 4 $mol\,l^{-1}$ HNO_3, the extent of dissolution inhibition increase in the succession $H_2SO_4 > HCl > HClO_4 > H_3PO_4$; which is the same order as revealed from the result of salt additions.

The corrodibility of lead in the absence or presence

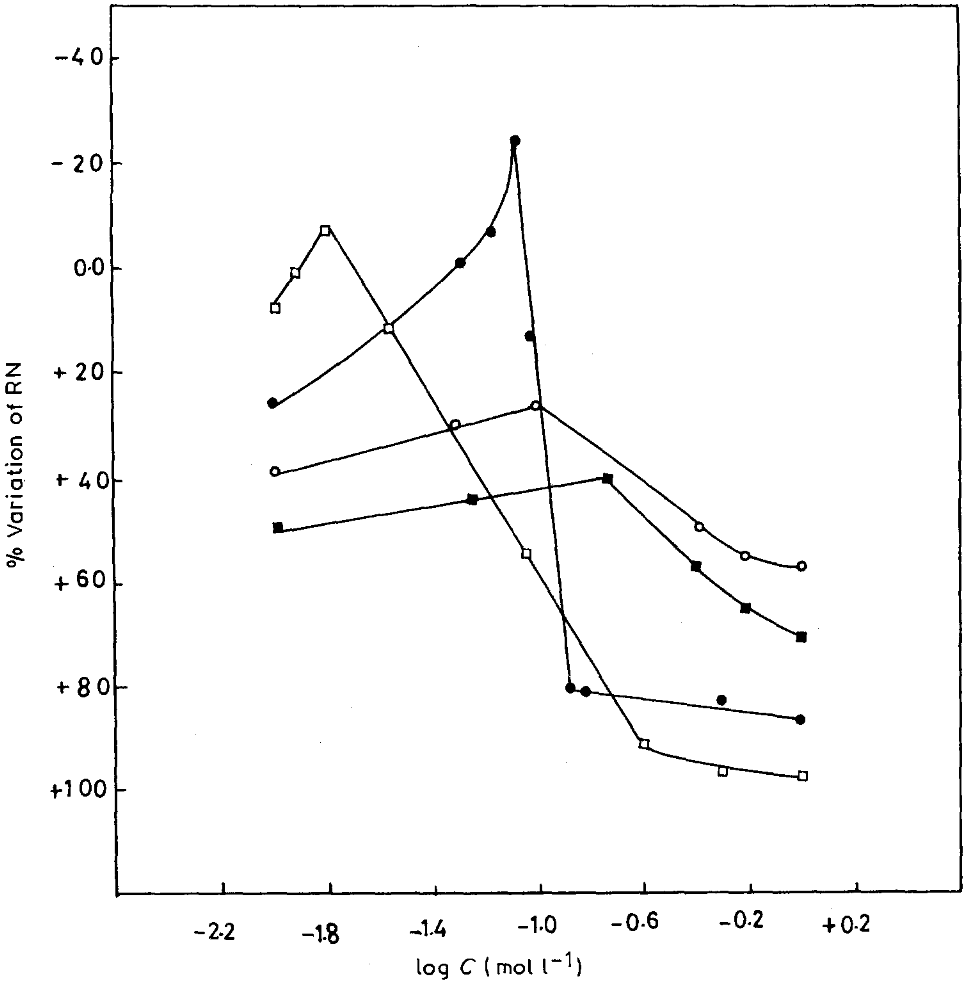

Figure 9 The percentage variation of RN with the concentration of various inorganic acid additives of Pb in 4 $mol\,l^{-1}$. HNO_3 (○ H_3PO_4, ■ $HClO_4$, ● HCl, □ H_2SO_4).

TABLE I Efficiency of corrosion inhibition, as determined from thermometric and weight loss methods: additive concentration 0.25 mol l^{-1}; positive value indicates inhibition, negative value acceleration of corrosion.

Additive	Inhibition (%)	
	Thermometric	Weight loss
Sulphuric acid (H_2SO_4)	+ 92.0	+ 89.9
Sodium sulphate (Na_2SO_4)	+ 88.5	+ 84.3
Hydrochloric acid (HCl)	+ 83.0	+ 81.4
Sodium chloride (NaCl)	+ 82.5	+ 78.0
Perchloric acid ($HClO_4$)	+ 43.5	+ 41.8
Sodium perchlorate ($NaClO_4$)	+ 42.0	+ 40.6
Orthophosphoric acid (H_3PO_4)	+ 41.5	+ 38.0
Sodium dihydrogen Orthophosphate (NaH_2PO_4)	+ 24.5	+ 21.3
Sodium nitrate ($NaNO_3$)	+ 18.5	+ 15.0
Sodium nitrite ($NaNO_2$)	− 30.0	− 27.0
Hydrazine (H_2N-NH_2)	− 40.0	− 38.0

of additives in 4 mol l^{-1} HNO_3, as established by the thermometric methods, was compared with that determined by the weight-loss technique. The additives examined are arranged in order to increase inhibition efficiency (or in the case of $NaNO_2$ and hydrazine accelerating effect) in Table I together with inhibition percent I obtained by the weight-loss method

$$I = [1 - (W_{\text{add}}/W_{\text{free}})] \times 100 \qquad (10)$$

where W_{free} and W_{add} are the weight losses in free solution and with additive, respectively. The agreement between the results of corrosion assessment by the two independent techniques is quite evident.

The same ranking of inhibitive action is thus obtained by weight-loss and thermometric measurements, and both techniques also show that only two of the additives accelerate corrosion. These results indicate that the thermometric method can be recommended as a reliable technique for the rapid evaluation of the corrosion of lead.

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