Chemical Characteristics of Water Influencing Corrosion

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INTRODUCTION

Metals immersed in water tend to corrode because of their thermodynamic instability. Natural waters contain dissolved solids and gases and sometimes colloidal matter ; all these may affect the corrosive properties of the water in relation to the metals with which it is in contact. The effect may be either one of stimulation or one of suppression and it may affect the cathodic or the anodic reaction. Some metals form a natural protective film in water and the corrosiveness of water to these metals depends on whether or not the dissolved material it contains assists in the maintenance of a self-healing film. The metals most commonly used for water systems are cast iron and mild steel. The concepts presented here are a review of the information available in the literature on the corrosive behaviour of different waters towards cast iron and mild steel.

TYPES OF CORROSION

The types of corrosion from the point of view of water are—(*i*) general corrosion, and pitting and (*ii*) caustic pitting ; while the former is a major problem of concern to those involved with design and operation of water utilities, the latter is of less significance as it is likely to occur only when mild steel is stressed in tension above the yield point and comes in contact with strong caustic solutions. The present discussion is limited only to the general corrosion and thus to the corrosive behaviour of waters.

General Corrosion and Pitting

The modern and now well established view of corrosion is that the process is almost without exception electro-chemical in character. Most of the cases of corrosion can be explained in terms of the formation of electrodes and the concomitant reactions which the flow of current across such zone induces. The commonly occurring reactions at the electrodes are shown below :—

$$\begin{aligned}
&\mathrm{Me} + p\mathrm{L}^{-m} &&= \mathrm{Me}\,(\mathrm{L})_p^{n-pm} + n\mathrm{e}^- &&(1)\\
&2\,\mathrm{H}^+ + 2\,\mathrm{e}^- &&= \mathrm{H}_2 &&(2)\\
&\mathrm{O} + 2\,\mathrm{H}^+ + 2\,\mathrm{e}^- &&= \mathrm{H}_2\mathrm{O} &&(3)\\
&\mathrm{H}_2\mathrm{O} + 2\,\mathrm{H}^+ + 2\,\mathrm{e}^- &&= 2\,\mathrm{H}_2\mathrm{O} &&(4)\\
&\mathrm{O}_2 + 4\,\mathrm{H}^+ + 4\,\mathrm{e}^- &&= 2\,\mathrm{H}_2\mathrm{O} &&(5)\\
&\mathrm{O}_2 + 2\,\mathrm{H}_2\mathrm{O} + 4\,\mathrm{e}^- &&= 4\,(\mathrm{OH})^- &&(6)
\end{aligned}$$

The reverse reactions often do not occur at appreciable rates at corroding metals and for simplification they are not considered.

These reactions are termed anodic or cathodic depending upon whether electrons are gained or lost by the metal and like any other heterogeneous reactions occur in successive steps. The tendency of a metal surface to give up positively charged metal ions to the water leaving electrons behind is represented by equation 1. Such a process should normally come to a halt owing to the potential difference built up between the metal and water. The reaction can continue further with dissolution of metal, only if the electrons are withdrawn from the metal. Such a withdrawal of electrons is effected in the 'hydrogen-evolution' type of corrosion (eq. 2) and the 'oxygen absorption' type of corrosion (eq. 6). In both types of corrosion, the metal is dissolved away at the points where the reaction (1) is taking place. At adjacent points, hydrogen is evolved with a reduction in acidity or oxygen is reduced to hydroxyl ions with a consequent increase in alkalinity. The requirements of the two types of corrosion thus differ. The hydrogen ions needed for the hydrogen evolution type are always present in the water but the oxygen absorption type cannot proceed without a continuing supply of dissolved oxygen.

Corrosion Rates

Progress of corrosion as measured by the determination of weight loss of a specimen that were exposed to different time intervals show that the corrosion reaction rate appears to proceed to a steady state in which corrosion rate becomes constant. The inhibitive properties of natural coatings that are formed on the surface by water are indicative of the rate of approach to this steady state. Stumm[1] states that the initial rate of corrosion is related to the salinity of the test waters and dissolved oxygen concentration. This indicates that in the early phases of corrosion process, the supply of dissolved oxygen to the cathodic regions of the surface and the electrolyte concentration control the rate of corrosion.

LANGELIER INDEX

It is commonly accepted belief that the corrosive behaviour of a natural water is influenced predominantly by pH and calcium carbonate saturation, frequently referred to as Langlier Index. To mitigate or reduce corrosion, it is conventional to increase the calcium and alkalinity content by adding lime. This is, however, an over-simplified concept and is not in general agreement with practical experience. The author is, therefore, of the opinion that the design of treatment for corrosion control should be based on an experimental evaluation rather than on any generalised concepts.

The tendency of a natural water to deposit calcium carbonate can be expressed by the reversible reaction :

$$Ca^{++} + HCO_3^- = CaCO_3 + H^+ \qquad (7)$$

The mathematical relationship between carbondioxide, calcium bicarbonate, and calcium carbonate has been studied by several workers, including Langelier. The simpler form of Langelier expression is based on the solubility product of reaction[7] for $CaCO_3$ (Ks) and the second ionisation equilibrium constant of carbonic acid (K_2)

$$pH_s = (pK'_2 - pK'_s) - \log (Ca^{++}) - \log (AlK) + \log x \qquad (8)$$

The expression can also be written as

$$pH_s = pK'_2 - pK_s' + pCa^{++} + PAlK - px \qquad (9)$$

(Ca^{++}) = concentration of calcium ions in moles per litre.
(Alk) = titrable alkalinity in equivalents per litre.
(H_s^+) = hydrogen ion concentration at a hypothetical saturation with calcium carbonate.

K'_2 = second ionisation of constant calcium carbonate corrected for activity.
K'_s = ion product of calcium carbonate, corrected for activity.

$$x = \left\{ 1 + \frac{2K'_2}{(H_s^+)} \right\}$$

This simple form does not apply to pH values over 9.0 and high salinities affect its accuracy. The term $(pK'_2 - pK'_s)$ is a function of temperature and ionic strength. Much useful information can be obtained from this equation in an analysis of a given water at a constant temperature[4]. The difference between the actual pH and saturation pH is referred to as saturation index of water (S.I.). The index is used as a practical measure of the tendency of a water to deposit $CaCO_3$. If the S.I. is positive, the water will be supersaturated with $CaCO_3$ whereas if it is negative the water will be aggressive to calcium carbonate. It is, however, not possible to predict the amount of $CaCO_3$ deposited by the equilibrium conditions alone because, the relative rates of $CaCO_3$ precipitation from solutions saturated with $CaCO_3$ control it and $CaCO_3$ has a strong tendency to remain supersaturated in solution. Calcium carbonate solution can be preserved for years in the absence of nuclei for crystalisation.

INFLUENCE OF pH AND HYDROXYL IONS

The pH of natural waters is rarely outside the narrow range of 6.0 to 9.0. High values at which the corrosion is suppressed and low values at which the gaseous hydrogen evolution occurs, are not often found in natural waters. According to weight loss measurements, steel corrodes at approximately the same rate throughout the range of pH found in natural waters. The form which the corrosion takes is, however, affected by pH[4].

In a water with a pH of 8.4, the deposition of $CaCO_3$ in 5 days was 213 mg per square decimeter of iron surface. In an identical experiment using stainless steel specimens, practically no deposition of $CaCO_3$ was noticed at the end of the test period though the saturation index was +1.65 pH unit[1]. This effect shows the influence of corrosion reaction at the iron surface. The shift of pH values which occurs near the cathodic regions of corroding surface is also demonstrated by the fact that even at negative saturation indexes, appreciable amounts of calcium carbonate are precipitated.

The hydroxyl ions may exert corrosion stimulating as well as corrosion mitigating influence. One must consider this in discussing the effect of pH upon corrosion. The solubility of corrosion products as well as the solubility of $CaCO_3$ decreases with increase in hydroxyl ion concentration. One would assume that corrosion rates decrease with increased pH on this aspect alone. It should be considered, on the other hand, that the buffer capacity of natural water decreases for a given alkalinity appreciably with increased pH upto **8.4**. An increase in buffer capacity associated with a decrease in pH should lead to decrease in corrosion rates. The optimum pH for a given water at which the corrosion can be lowest would thus be defined by a net balance of these opposing effects. Different pH optima can be observed for waters of different mineral composition, the action of many corrosion inhibitors being strongly pH dependent.

Buffer Capacity

Buffer capacity may be expressed in terms of the amount of strong base required to produce a unit change in pH of the solution. It is low with low alkalinity soft waters. More significant increase in pH value near the metal surface is produced as a result of corrosion reaction resulting in precipitation of more calcium carbonate. Ryznar[5] points out that the deposition of $CaCO_3$ is more dependant on the pH than on saturation index. He proposes a stability index, $S = 2\,pH_s - pH$, which is believed to be a better indication of the expected amount of $CaCO_3$ deposition. This empirical index gives more importance to saturation pH than does Langelier Index. To a certain extent the saturation pH is a function of buffer capacity.

The corrosion rates are usually lower on hard waters than in soft waters. In spite of the greater deposition of $CaCO_3$ in soft waters, there can also be higher corrosion rates[1]. From this it appears that thickness of $CaCO_3$ alone is not sufficient for corrosion properties. A possible inference is that the crystalline state of the precipitated $CaCO_3$ and iron is also of importance. In a soft water, $CaCO_3$ is deposited in a more amorphous state than in hard water and the precipitated iron salts show a different structure than in raw water.[6] Another inference is that in the presence of greater amounts of electrolytes, colloidal iron hydroxide is more rapidly precipitated on the iron surface because of a reduction in pH.

The buffer capacity (β) of a natural water can be expressed as :

$$\beta = 2.3 \left[\frac{(H^+)\,(Alk)}{(H^+) + 2K_2} \left\{ \frac{(H^+)}{(H^+) + K_1} + \frac{K_2}{(H^+) + K_2} \right\} + (H^+) - (OH^-) \right] \quad - (10)$$

Where β is in equivalents per pH unit, (Alk) in equivalents per litre, and (H^+) and (OH^-) concentration are in moles per litre. K_1 and K_2 are 4×10^{-7} and 4.6×10^{-11} respectively.

Alkalinity

The solubility of ferrous and calcium ions is reduced and the buffer capacity is increased with an increase in alkalinity content. Even under favourable pH conditions, the coatings formed in low alkaline pH waters cannot be protective against corrosion. The inhibitive effect of bicarbonate and carbonate ions is indirect, as these ions are not specifically adsorbed on the metal surface though they are readily hydrated.

The finding that calcium in the presence of alkalinity, regardless of pH or saturation index, is an effective inhibitor is not consistent with theory and experience[7,8,9]. The basic corrosion reaction is conducive to the formation of $CaCO_3$ at the point of corrosion even though the water environment is below saturation with respect to $CaCO_3$. The rate at which such deposition may take place is increasingly altered by higher velocities and is improved by pH adjustments to or above the saturation pH for $CaCO_3$[10,15]

DISSOLVED OXYGEN

In the natural waters which are saturated with $CaCO_3$ the dissolved oxygen may under certain conditions have no accelerating influence on the corrosion rate because it also promotes the film formation. There is no evidence in the literature to indicate to the contrary that waters relatively high in dissolved oxygen will not produce better protective coating on corrodible ferrous metals than the same waters which are low in dissolved oxygen[11]. The dissolved oxygen has a dual influence on the corrosion process. Firstly, it is a good cathodic depolariser, increasing the rate of cathodic process and when the latter is the controlling factor, the corrosion rate. Secondly, dissolved oxygen increases the electrode potential. Cohen[9] has shown that the oxidising agent is frequently necessary for the formation of an anodic protective film. High concentrations of dissolved oxygen thus lower the probability of corrosion by improving the anodic films[1]. It is considered that dissolved oxygen is adsorbed on the surface of the iron and takes part in the formation of a protective film.

INFLUENCE OF CHLORIDES AND SULPHATES

It is well established that the passive anodic film comprises 50-200Å thin anhydrous y-Fe_2O_3. This is destroyed by chloride and to a smaller extent by sulphate. These ions are anodic corrosion stimulants and increase the anodic area. Larson and Skold[12] determined the relative influence of chloride and sulphate ions on tuberculation. Upto 25 mg/l chlorides, 30 mg/l sulphates, 85 mg/l calcium hardness, and 120 mg/l alkalinity no significant difference with regard to corrosion rate at tuberculation was observed. Chloride and buffer capacity are antagonists and Larson and Skold[12,13,14,15,16] have determined the corrosive inhibitive relationship for various equivalent rates of bicarbonate chloride for given pH values.

Chloride—Alkalinity Ratio

Larson and Skold[12] have determined the effect of chloride/alkalinity ratio on the corrosion of machined cast iron specimen in waters with an alkalinity of 125 mg/l and calcium hardness of 30 and 85 mg/l. The pH was regulated in the experiments to maintain a zero saturation index. The experiments clearly indicated that the relationship of increased corrosion rate to increased chloride/alkalinity ratio is minimised within 30 days when the water is stabilised with $CaCO_3$. The beneficial effect of higher calcium hardness is shown even though the pH is lower.

Iron and Manganese

The small amount of deposit from these ions is not likely to have much effect on corrosion although there is always a possibility that attack will occur under the sludge deposits. Most iron bearing waters contain substantial amounts of carbonic acid which is more troublesome.

Organic matter

Some of the worst corrosive effects in soft water are attributed to a rather wide group of organic acids abstracted from peat. Such waters usually have low pH values and are often discoloured. They effect ferrous metals appreciably and corrosion control is rarely achieved by pH correction of such waters[4].

CONCLUSIONS

It is obvious that no single test procedure will provide data which can characterise all chemical aspects of water leading to corrosion and its consequences. It is important to realise that the scale deposition is frequently the indirect result of corrosion. It has been demonstrated that waters significantly oversaturated with $CaCO_3$ do not deposit at non-corroding surfaces. On the other hand $CaCO_3$ can deposit from unsaturated waters on corroding surfaces. The local increase in pH at cathodic neighbourhood is responsible for this.

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DISCUSSION

Mr. A. G. Nene (Shriram Chemical Industries, Kota)

We face considerable corrosion problem in our Ammonia/Urea circulation water circuit:

450-500 M^3/hr fresh water is supplied from water softening plant. This passes through the compressor jackets and cools air for compressor motors. The hot water is diverted to hot well of cooling tower. At hot well, 10,000 M^3/hr of hot circulating water is received after cooling from various condensers of ammonia and urea plants. Hot water is cooled in cooling tower and is recirculated as above.

From the circulation water, 350 M^3/hr of water is drained as the water is contaminated and unfit for reuse.

	Fresh water	Discarded water
TDS of water	120 ppm	140-160 ppm
Free chlorine	Nil	0.2 - 0.3 ppm

Average temperature rise at hot end is 5-6°C.
With this set up we are facing the following problems:

1. Deposition of algae on open heat exchangers.
2. Slime deposits inside a condenser.
3. Deposition of scale at hot end of few condensers where temperature rise is 15-20°C.
4. Small oxidation type corrosion marks at heat exchangers at cold end of water.
5. Sometimes water pH goes to 9 and above due to entry of ammonia/urea at cooling towers.
6. Insects get pulled at cooling towers and choke heat exchanger tubes.

Material of construction for most of the condensers and tubes is mild steel. Few are of stainless steel.

Mr. K. R. Bulusu (CPHERI, Nagpur)

The observations made by Shri Nene are quite interesting. There will be corrosion at points due to the differences in the concentration of oxygen. When dissolved oxygen is high, the hydroxyl ion concentration is appreciable and consequently the precipitating tendency is more. Whereas at the low dissolved oxygen levels, the amount of hydroxyl ion concentration is less. It is such difference in the dissolved oxygen concentration that gives rise to what is referred to as reduction type corrosion. Corrosion is not completely preventable. It can be reduced by using some inhibitors. Calcium carbonate is one such inhibitor. It forms a layer at the surface and reduces corrosion. We have other types of inhibitors like phosphates and chromates.

Dr. K. S. Rajagopalan (CECRI, Karaikudi)

I am not going to dispute many of the statements made in the lecture about the dynamic tendency of iron to get into solution in water. There are two types of reactions taking place, the one being oxygen reduction. The aqueous corrosion is distinct from oxidation in the air in the sense that the two types of reactions take place at different times and different conditions. Coming to corrosion by water, the speaker referred to the important role played by oxygen. In industrial waters, the solubility of oxygen is simply a function of temperature. As the speaker pointed out, you have something like 6-9 mg/lit. of oxygen. Under extraordinary conditions, there may be considerable enrichment of oxygen in water. But you have to generally deal with about 6-9 mg/1 of oxygen. This would mean that if oxygen reduction is important for corrosion, the corrosion rates should more or less be within a small range. We have tested at least 15-20 samples of water from different parts of the country where concentration of total impurities, which can play an important part in the corrosion process, varied from 40 mg/1 to 30,000 mg/1 or 40 ppm to 130 ppm. We found that corrosion is governed by the solubility of the oxygen in water in the sense that all the water had the corrosion rate of 7-14 $mg/dm^2/day$.

Another important point discussed in this meeting deals with Langlier Index which, I would like to point out, was not evolved to give a solution to the problem of corrosion. This index only shows whether water is capable to throw out calcium carbonate or dissolve calcium carbonate. The prevention of corrosion is due to secondary effect of formation of a layer of $CaCO_3$. If Langlier index is positive the corrosion rate is likely to be reduced. But this does not necessarily occur all the time. We have observed that even when the index varied from -1.5 to $+1.8$, the water had corrosion rates of 7-14 $mg/dm^2/day$. So far as the supply of water in this country is concerned and the ambient temperature involved, the kind of $CaCO_3$ that is thrown out does not form a barrier type of film. Values of corrosion rate obtained in industrial water vary in the same range as the corrosion rate obtained in poor solution of chloride. The usefulness of Langlier index comes into use only when there is an egg-shell type of film formed, i.e., if certain type of organic compounds are present. Otherwise in the presence of chloride the kind of incrustation that is formed is not suitable to reduce corrosion. The solution to the problem therefore lies in adding substances to water which can form an invisible film.

MR. L. M. AGARWAL (Sudbury Laboratory of India, Calcutta)

For this kind of corrosion problem we feel that the simplest solution will be to have some inhibitors to form a micro-film type of layer on the metal surface, thereby not allowing the water to come in contact with the metal.

MR. S. G. KRISHNAN (NOCIL, Thana)

The variation in chloride content and pH of the water at the metal/solution interface, leads to non-uniformity in the protective film formation. I agree that theoretically oxygen is one of the main criteria for corrosion but it is not possible to avoid this as all the cooling towers in the world are saturated with oxygen. Our aim should be to develop such inhibitors which will take care of chloride concentration, pH fluctuations and still give a uniform protective film. We have done some work in the laboratory and found that zinc, sodium polyphosphate combination eliminates pitting type of corrosion in places where extremely soft water is used.

MR. K. R. BULUSU (CPHERI, Nagpur)

Inhibitors will provide protective film over the anodic and cathodic point and will reduce corrosion. When we consider inhibitors, some of their important aspects have to be kept in view. When chromate is used as inhibitor its toxicity should be borne in mind. Phosphates are good inhibitors provided we can keep them in solution and it is difficult to keep them in solution in the natural waters. The molecularly dehydrated phosphates have the ability to stabilise an otherwise unstable water and even in the presence of small amounts of these phosphates, a water that would ordinarily form a heavy deposit of $CaCO_3$ remains stable for extended period of time.

Having brought out the limitations of Langlier Index, we have to consider an alternative. The alternative, as it appears today, is Ryzner or Saturation Index. The Saturation Index is a function of pH and saturation pH of calcium carbonate (pHs.) It is given as 2 pHs — pH. Knowing the pH of saturation at a given temperature, we will be able to evaluate the saturation index. The Ryzner or Saturation Index is always positive. The implication is, if the numerical value of the index is less than or equal to 6, we refer the water as definitely "scale forming". If the index is greater than or equal to 8, the water is definitely said to be corrosive. When the index is between 6 and 8, it means that the system cannot be predicted with confidence.

By adopting Ryzner Index we can, therefore, obtain a definite indication of the nature of the water. The usefulness of this index can be seen more clearly by assuming that there are two waters of the following characteristics.

Water (a) at 75°C — actual $pH=6.5$, $pHs=6.0$

Langlier $Index=pH-pHs=6.5-6.0=+0.5$

Water (b) at 75°C — actual $pH=10.5$, $pHs=10.0$

Langlier $Index=pH-pHs=10.5-10.0=+0.5$

From the Langlier Index, it might be predicted that both waters would be equally scale forming. Actually (a) would be scale forming (b) would be quite corrosive.

Such a possibility of misinterpreting can be overcome by using Ryzner Index. This index is not only an index of calcium carbonate saturation but is also of quantitative significance.

Using the waters (a) and (b) described above, the following values would be obtained for the Ryzner Index.

	Langlier Index	Ryzner Index
Water (a)	+0.5	−5.5
Water (b)	+0.5	−9.5

This shows that water (a) with an index of 5.5 is scale forming and will give an appreciable amount of calcium carbonate scale. Water (b) on the other hand has an index of 9.5 and can be severely corrosive at higher temperature.

DR. M. M. LOTLIKAR (NOCIL, Thana)

The inhibition by phosphate as well as zinc chromate is extremely reliable. As far as chromates are concerned, we always found that you have to use 200 ppm of chromates. But still this gives certain problem and it is very costly even in India. Therefore, a cheaper system will be to go for phosphate. Finally, we have developed a system to use polyphosphate which is cheaper.

MR. L. M. AGARWAL (Sudbury Laboratory of India, Calcutta)

With the addition of phosphate will there be no likelyhood of increase in algae growth?

DR. M. M. LOTLIKAR (NOCIL, Thana)

To solve the problem of algae growth we used chlorine. If the chlorine is sufficient that means a little more free chlorine than O ppm then all the bacteria is killed.

DR. A. M. TRIVEDI (Gujarat University, Ahmedabad)

I think that the discussion has made these two points clear that if a film is formed in such a way that it can form a barrier it is all right. Therefore, we have not only to avoid corrosion but should also avoid building up of massive incrustation. What we want is a sufficiently thin film. But in such a case there is a danger of localised corrosion.

We have anodic and cathodic inhibitors which help in the formation of thin visible film. We have a combination of both anodic and cathodic inhibitors, zinc phosphate organic phosphate so that we avoid corrosion as well as incrustation. The problem of pitting is more or less over come by using this combination. When we use phosphate of zinc and calcium then only we have a better solution to the corrosion problem.

MR. S. N. SHUKLA (Public Health Dept., Nagpur)

After a cast iron pipeline is laid underground, there are cases of bursting of pipeline. We are trying to find out the reasons. When the pipelines are laid they are subjected to impact and some cracks are developed which afterwards lead to bursting. There may also be some invisible cracks in the pipeline before laying which remain unnoticed.

MR. L. P. BORKAR (Bombay Municipal Corporation, Bombay)

We have also experienced a similar problem in a pumping sewage line of 1000 feet length, laid 8 years back. At the inlet of the pipeline there was a burst — a regular crack. A similar crack was detected earlier.

DR. A. K. LAHIRI (Corrosion Advisory Bureau, Jamshedpur)

It is probable that the problem referred by Mr. Borkar is a case of failure due to vibration caused by either cavitation or other mechanical causes. In presence of corrosive agent the fatigue type failure may be accelerated.

MR. L. P. BORKAR (Bombay Municipal Corporation, Bombay)

We have experienced similar problem and found it to be due to hammer effect. What we did was, we prepared a buffer tank about 300 feet high and there is not a single breakage for the last 2½ years. In this connection (1) design and laying conditions and (2) hammer effect are important where water hammer effect occurs due to pressure changes taking place along the pipes. It is necessary to have a proper study of the flow in the pipe to have a solution.

MR. V. M. SHIDHAYE (Bombay Municipal Corporation, Bombay)

The failure of joints in cast iron pipes is mainly due to unequal settlement. After excavation and laying the pipe when the trench is refilled there is always a gap left below the pipe. There is unequal pressure developed due to settling of soil and that is the reason why joints give way.