CRYSTALLIZATION FOULING

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Contents

Summary

The formation of crystalline deposits on the heat transfer surfaces is one of the main problems in thermal desalination processes. Precipitation starts once the concentration of a certain salt exceeds its equilibrium solubility. Equations for this condition are provided in this chapter. Depending on operating conditions, the deposition process can be diffusion or reaction controlled, or a combination of both. Based on a careful literature survey, the effect of operating parameters, such as flow velocity, surface temperature and salt concentration, are described. Correlations are provided for the prediction of deposition rates for heat transfer due to forced convection and/or subcooled flow boiling. Finally, crystallization fouling in shell and tube, plate and frame and fin-tube heat exchangers is discussed.

1. Introduction

Water is generally classified as a universal solvent, since almost everything is soluble in it to some extent. Because of its dissolving power, water can leach significant concentrations of salts as well as other materials with which it comes in contact. Variations of temperature, pressure, pH and the relative concentrations of other substances in solution control the solubility of a given salt in water. Once the solubility of dissolved material is exceeded some of the dissolved salts will precipitate out to form deposits.

In processes such as evaporation of saline waters or sea waters, various salts will precipitate in a certain order depending on operating temperature, pressure, ionic strength of solution, etc. The solubility of most salts increases with increasing temperature, and as a rule, these salts do not crystallize unless their concentrations are extremely high. Scale deposits are formed from those salts whose solubilities are generally limited and in most instances decrease with increasing temperature. The principal constituents that cause scaling problems in seawater and most industrial evaporators are calcium sulfates (gypsum, hemihydrate and anhydrite) and calcium carbonate. Theoretically, the deposition of calcium carbonate can be controlled by maintaining a slightly acidic pH. Calcium sulfate, however, is not significantly affected by pH and tends to precipitate in various forms once the water becomes supersaturated. Next to corrosion, the major limiting economic factor in evaporation processes is the degree to which seawater or industrial fluids can be concentrated before calcium sulfate scale occurs. For example, seawater becomes saturated with respect to calcium sulfate anhydrite when it is evaporated at 100°C to two-thirds of its original volume (Langelier et al. 1957).

Therefore, knowledge of the saturation concentrations of salts and salt mixtures under various operating conditions is essential to predict the tendency of water to form scale. Furthermore, almost all researchers express the rate of crystallization of these salts with respect to the degree of supersaturation driving force. Consequently, accurate knowledge of the saturation concentration of these salts is a prerequisite for the prediction of their rate of deposition on heat transfer surfaces.

2. Chemical Fundamentals

2.1. $CaCO_3$ Solubility in Water

Calcium carbonate crystals exist in three forms, namely aragonite, calcite and vaterite. Since all three forms of this salt have an inverse solubility, their solubilities in water will decrease as temperature increases, e.g. at a heated surface. This trend is illustrated in Figure 1 where the solubilities of various calcium carbonates are plotted as a function of temperature (Plummer and Busenberg 1982). Recently Najibi et al. (1997) have shown that in subcooled flow boiling, more than 99 per cent of calcium carbonate deposits in the form of aragonite, the solubility of which has been studied in detail by Plummer and Busenberg (1982). The solubility product of calcium carbonate is defined as:

$$K_{sp}=\left[Ca^{2+}\right]\left[CO_3^{2-}\right] \qquad (1)$$

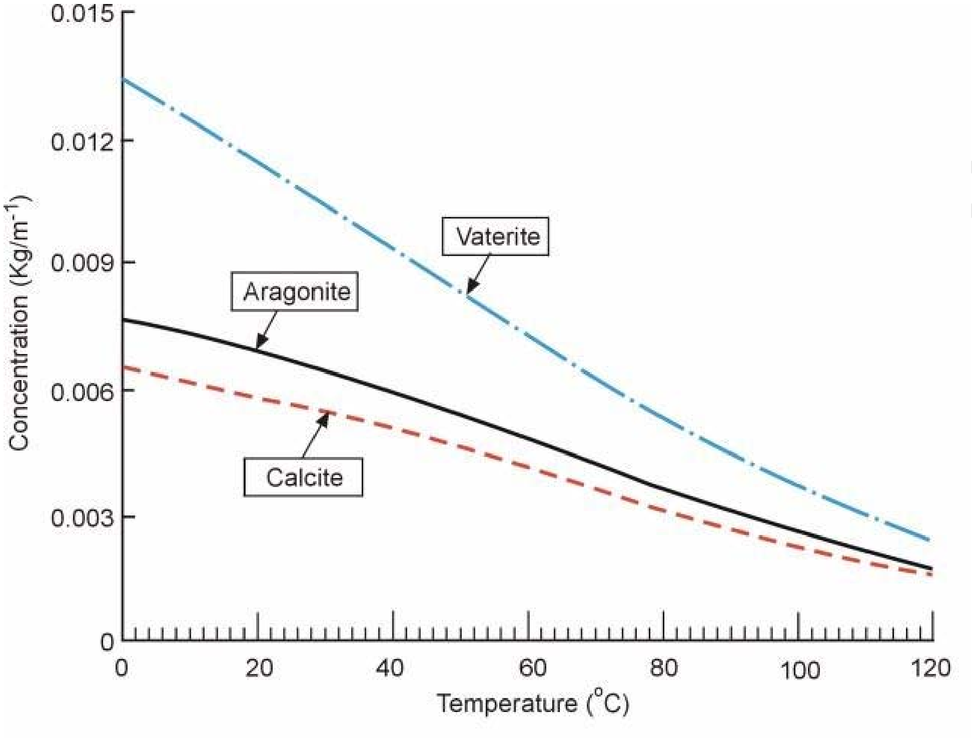

Figure 1. Solubility of calcium carbonate in water as a function of temperature.

Therefore, in the absence of common ion effects, the solubility of calcium carbonate (aragonite) in water becomes the square root of the solubility product, K_{sp}, which is given by the following equation (Plummer and Busenberg 1982):

$$\log(K_{sp})=-171.9773-0.077993T+\frac{2903.293}{T}+71.595\log(T) \qquad (2)$$

In eq. (2), T is in degrees Kelvin and K_{sp} in molar units.

2.2. Solubility of calcium carbonate for variable pH

Calcium carbonate precipitation is usually caused by pressure drop releasing CO_2 from bicarbonate ions (HCO_3^-). The effect of partial pressure of CO_2 on the pH of water is shown in Figure 2 at three different operating temperatures (Cowan and Weintrilt 1976). When CO_2 is released from water, the pH increases, the solubility of dissolved carbonate decreases and consequently the more soluble bicarbonates are converted to less soluble carbonate. As an illustration, the loss of 100 mg l^{-1} of bicarbonate deposits 28.6 lb/1000 bbl of water in calcium carbonate.

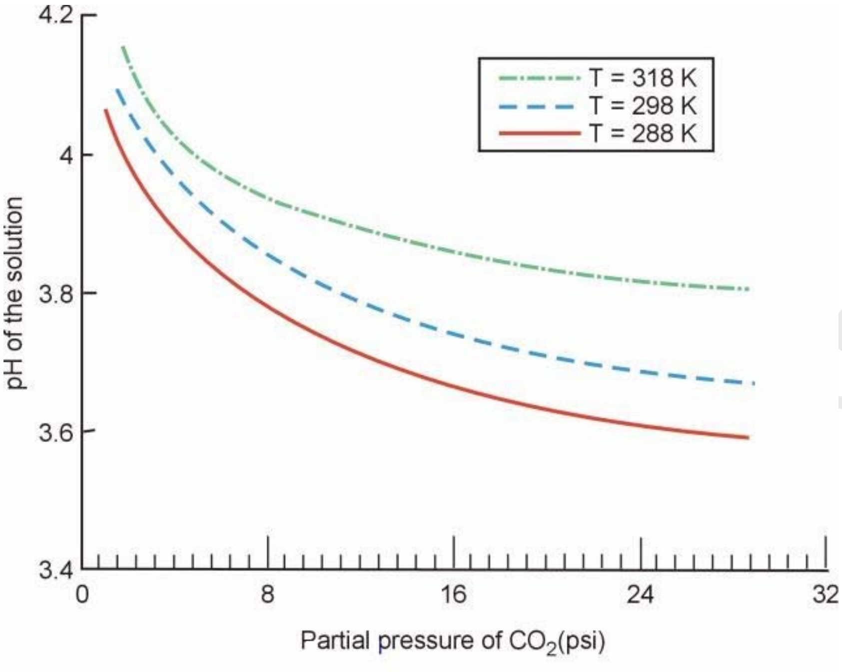

Figure 2. Effect of CO_2 partial pressure on pH of water.

The following reactions take place in aqueous solutions of calcium carbonate:

$$CaCO_3(s) \Leftrightarrow Ca^{2+} + CO_3^{2-} \tag{3}$$

$$CO_3^{2-} + H_2O \Leftrightarrow HCO_3^- + OH^- \tag{4}$$

$$HCO_3^- + H_2O \Leftrightarrow H_2CO_3 + OH^- \tag{5}$$

$$H_2O \Leftrightarrow H^+ + OH^- \tag{6}$$

Thus the total carbon concentration is:

$$C_T = [HCO_3^-] + [H_2CO_3] + [CO_3^-] \tag{7}$$

The total alkalinity (TA) of the solution is given by:

$$[TA] = [HCO_3^-] + 2[CO_3^{2-}] + [OH^-] + [H^+] \tag{8}$$

In order to take into account the effect of pH on the solubility of calcium carbonate, the distribution constants of carbonic acid should be taken into consideration:

$$K_1 = \frac{\left[H^+\right]\left[HCO_3^-\right]}{\left[CO_2\right]} \qquad (9)$$

$$K_2 = \frac{\left[H^+\right]\left[CO_3^{2-}\right]}{\left[HCO_3^-\right]} \qquad (10)$$

where, K_1 and K_2 are the first and second molar distribution constants of carbonic acid, which are functions of operating temperature and can be calculated from the following equations (Plummer and Busenberg 1982):

$$\log\left(K_1\right) = -356.3094 - 0.06091964T + \frac{21834.37}{T} + 126.8339 \log T - \frac{1684915}{T^2} \qquad (11)$$

$$\log\left(K_2\right) = -107.8871 - 0.03252849T + \frac{5151.79}{T} + 38.92561 \log T - \frac{563713.9}{T^2} \qquad (12)$$

Simultaneous solution of equations (8), (9) and (10) for the three different unknowns yields:

$$\left[CO_3^{2-}\right] = \frac{\left[TA\right] + \left[H^+\right] - \left[OH^-\right]}{2\left(1 + \frac{\left[H^+\right]}{2K_2}\right)} \qquad (13)$$

$$\left[HCO_3^-\right] = \frac{\left[TA\right] + \left[H^+\right] - \left[OH^-\right]}{\left(1 + \frac{2K_2}{\left[H^+\right]}\right)} \qquad (14)$$

$$\left[CO_2\right] = \frac{\left[TA\right] + \left[H^+\right] - \left[OH^-\right]}{\frac{K_1}{\left[H^+\right]}\left(1 + \frac{2K_2}{\left[H^+\right]}\right)} \qquad (15)$$

Thus, by knowing pH and TA of the solution, and the dissociation constants for carbonic acid, the concentrations of all species present in the solution can be calculated. Replacing the calculated concentrations and operating temperature in equations (1) and (2), the tendency of the solution for the crystallization fouling of calcium carbonate can be determined.

2.3. $CaSO_4$ Solubility in Water

The solubility of calcium sulfate in water is shown in Figure 3 as a function of operating temperature (Landolt-Bornstein 1985). At temperatures higher than 40°C, the solubility of calcium sulfate decreases with increasing temperature for the ordinary solid phases. Under normal operating conditions, the hottest portion of water is the layer next to the heat transfer surface. Any precipitation is, therefore, expected to take place here.

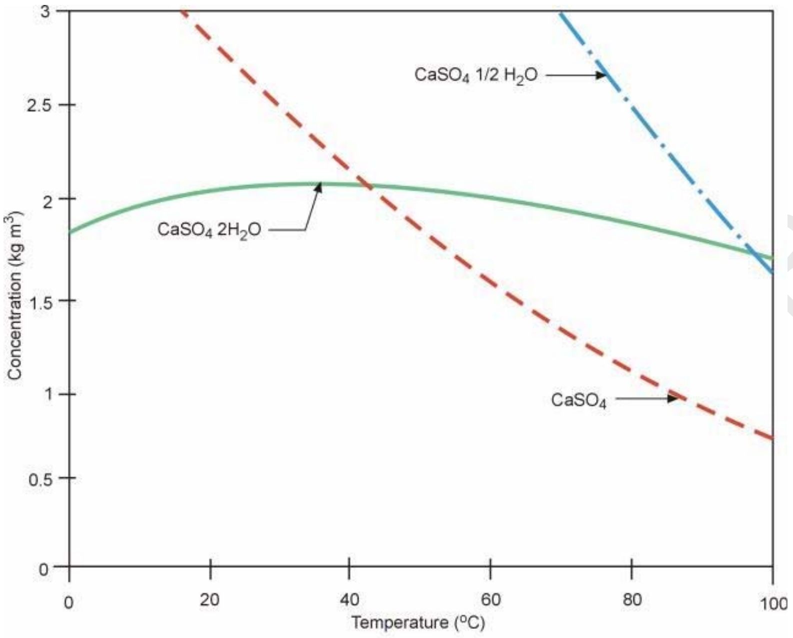

Figure 3. Solubility of calcium sulfate in water.

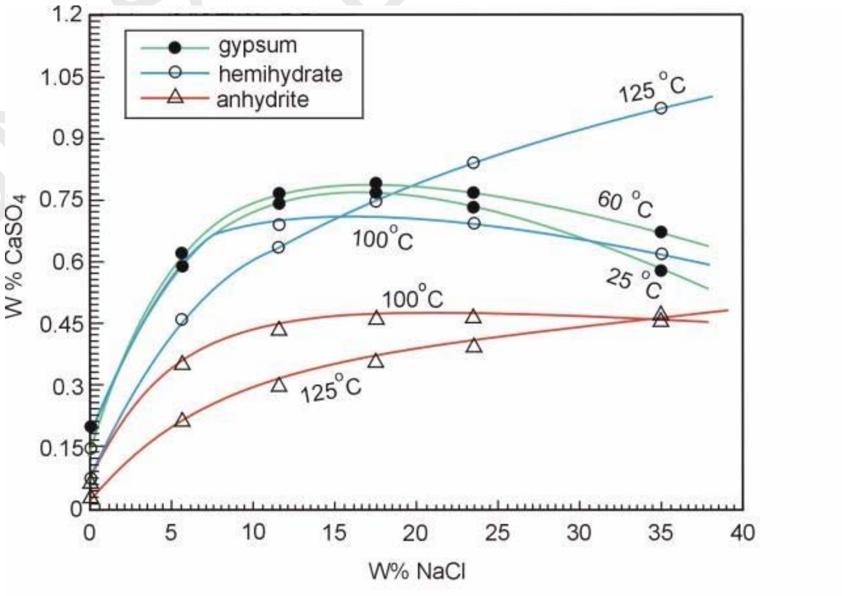

Figure 4. Effect of sodium chloride concentration on the solubility of calcium sulfate.

Seawater contains substantial amounts of other ions, especially sodium and chlorine. These ions are formed mostly from the dissolution of sodium chloride. The solubility of calcium sulfate is strongly affected by the presence and concentration of other ions in the system.

The solubilities of various calcium sulfates are shown in Figure 4 as a function of the sodium chloride concentration for different temperatures (Marshal and Slusher 1960). The solubility is increased with the concentration of sodium chloride, and decreased with increasing temperature. Most of the deposited calcium sulfate found in seawater desalination plants and in other industries is in the form of hemihydrate, the solubility of which has been studied in detail by Marshal and Slusher (1960). Najibi (1997), Najibi et al. (1997) by non-linear regression analysis of these data, developed the following correlation for the prediction of the saturation concentration of calcium sulfate hemihydrate as a function of ionic strength of solution composition and temperature.

$$C^{*} = 136\left(10^{a+bz}\right) \tag{16}$$

C* is in kg m^{-3} and parameters a, b and z are:

$$\left.\begin{aligned} a &= 2.047 - 0.01136T(K) \\ b &= -6.5832 + 0.0226T(K) \end{aligned}\right\} \tag{17}$$

$$z = \frac{\sqrt{I}}{1+1.5\sqrt{I}} \tag{18}$$

In these equations, I is the ionic strength expressed in kmol m^{-3}.

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