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WATER STABILIZATION IN WATER SUPPLY CIRCULATION SYSTEMS

СТАБІЛІЗАЦІЯ ВОДИ В СИСТЕМАХ ОБОРОТНОГО ВОДОПОСТАЧАННЯ

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Abstract. The aim of this work is to develop an algorithm for reagent stabilization of water in closed-loop water supply systems (CWSS) to prevent carbonate deposits on boundary surfaces and heat exchange, which is based on a physicochemical model of a carbonate water system (CWS) and allows determining the dose of reagents based on data from routine determination of recycled water (RW) parameters and CWSS operating mode. This approach avoids overdosage of reagents and discharge of excess RW into the environment, and if the RWSS is a recirculating cooling system (RCS) of thermal and nuclear power plants, it avoids emissions of excess fuel combustion products and a decrease in the efficiency of the plant due to deposits.
Methods. It is known that calcium carbonate is the main component of low-temperature deposits [1, p. 384]. In view of this, the article considers the hydrocarbonate subsystem of circulating water, the state of which is described by a stationary physicochemical model of water quality, which is formulated on the basis of:
1. The water-salt balance of the aquatic environment, taking into account changes in its volume.
2. Equations of parallel-sequential reactions reflecting transformations in circulating water.
3. Assumptions about the form of kinetic equations describing the crystallization of calcium carbonate, as well as the exchange of CO_2 between the atmosphere and the aquatic environment.
4. The law of conservation of the total amount of carbonates.
5. Conditions of electrical neutrality of the aquatic environment.
6. Taking into account the significant difference between the reaction rates in the hydrocarbonate system (HCS) relative to each other and the water turnover time in the system.
Results. Kinetic equations have been formulated for components that are neutral with respect to transformations in the water environment; we will call them tracers, and those that undergo changes as a result of physicochemical reactions in the water environment; we will call them forming deposits. Deposits arise as a result of the processes of mutual transformation of HCS components in the cooling water, therefore, a kinetic equation is formulated for the sum of carbonates in the cooling water, which has no restrictions on the constancy of the RCS volume and takes into account the processes of solid $CaCO_3$ formation and CO_2 degassing in cooling towers. For steady-state operation of the RCS, the kinetic equation for the sum of carbonates is converted into an equation for the rate of $CaCO_3$ release, which takes into account the change in the concentration of hydrocarbonate ions and CO_2 degassing. This equation allows us to estimate the effect of sulfuric acid dosing in the makeup water on the $CaCO_3$ precipitation rate and determine the acid dose sufficient to minimize $CaCO_3$ deposits. The concentration of hydrocarbonate ions and Ca^{2+} ions in the cooling water and makeup water is determined, which minimizes the rate of $CaCO_3$ precipitation at a given acid dose.
Practical value. The equation for the rate of $CaCO_3$ precipitation, which takes into account changes in the concentration of hydrocarbonate ions and CO_2 degassing in cooling towers, logically reflects the results of measures to stabilize the circulating water with acid, and allows obtaining the concentration of Ca^{2+} in the circulating and feed water that minimizes $CaCO_3$ deposition, as well as determining the proportion of feed water that should be treated with lime to minimize deposition, while avoiding overdose of reagents and damage to the environment. Moreover, the equations obtained allow, based on operational data on the quality of circulating water, to monitor the effectiveness of stabilization measures and adjust them in a timely manner.
Key words: circulating water; hydrocarbonate subsystem; minimization of $CaCO_3$ deposits; overdose of reagents.

Анотація. Метою даної роботи є розробка алгоритму реагентної стабілізації води в системах оборотного водопостачання (СОВП) для запобігання карбонатним відкладенням на граничних поверхнях та теплообміну, який базується на фізико-хімічній моделі карбонатної водної системи (КВС) і дозволяє визначати дозу реагентів на основі даних штатного визначення параметрів оборотної води (ОВ) та режиму роботи СОВП. Такий підхід дозволяє уникати передозування реагентів та скидання надлишкової ОВ у навколишнє середовище, а якщо СОВП є оборотною системою охолодження (ОСО) теплових та атомних електростанцій, то дозволяє уникати викидів надлишкових продуктів згоряння палива та зниження коефіцієнта корисної дії станції внаслідок відкладень.
Методи. Відомо, що карбонат кальцію є основним компонентом низькотемпературних відкладень [1, с. 384]. З огляду на це, в статті розглядається гідрокарбонатна підсистема оборотної води, стан якої описується стаціонарною фізико-хімічною моделлю якості води, яка формулюється на основі:
1. Водно-сольового балансу водного середовища з урахуванням змін його обсягу.
2. Рівнянь паралельно-послідовних реакцій, що відображають перетворення в оборотній воді.
3. Припущення про форму кінетичних рівнянь, що описують кристалізацію карбонату кальцію, а також обмін CO_2 між атмосферою та водним середовищем.
4. Закону збереження загальної кількості карбонатів.
5. Умови електричної нейтральності водного середовища.
6. Врахування істотної різниці між швидкостями реакцій в гідрокарбонатній системі (ГКС) відносно одна одної та часом обороту води в системі.
Результати. Сформульовано кінетичні рівняння для компонентів, які є нейтральними щодо перетворень в ОВ; ми будемо називати їх трасерами, та тих, які зазнають змін в результаті фізико-хімічних реакцій в ОВ; ми будемо називати їх накипінами. Відкладення виникають в результаті процесів взаємного перетворення компонентів ГКС в ОВ, тому формулюється кінетичне рівняння для суми карбонатів в ОВ, яке не має обмежень щодо постійності об'єму ОСО і враховує процеси утворення твердого $CaCO_3$ та дегазації CO_2 у градирнях. Для стаціонарної роботи СОВП кінетичне рівняння для суми карбонатів перетворюється на рівняння для швидкості виділення $CaCO_3$, яке враховує зміну концентрації гідрокарбонатних іонів і дегазацію CO_2. Це рівняння дозволяє оцінити вплив на швидкість осадження $CaCO_3$ дозування сірчаної кислоти у воду підживлення і визначити дозу кислоти, достатню для мінімізації відкладень $CaCO_3$. Визначається концентрація гідрокарбонатних іонів і іонів Ca^{2+} у ОВ і підживлюваній воді, яка мінімізує швидкість осадження $CaCO_3$ при заданій дозі кислоти.
Практична цінність. Рівняння для швидкості виділення $CaCO_3$, яке враховує зміни концентрації гідрокарбонатних іонів і дегазацію CO_2 в градирнях, логічно відображає результати заходів щодо стабілізації оборотної води кислотою, і дозволяє отримати концентрацію Ca^{2+} в оборотній і та підживлюючій водах, які мінімізують відкладення $CaCO_3$, а також визначити частку підживлюваної води, яку слід обробляти вапном, щоб мінімізувати відкладення, уникаючи при цьому передозування реагентів та шкоди навколишньому середовищу. Більше того, отримані рівняння дозволяють, на основі оперативних даних про якість оборотної води, контролювати ефективність заходів стабілізації та своєчасно їх коригувати.
Ключові слова: оборотна вода; гідрокарбонатна підсистема; мінімізація відкладень $CaCO_3$; передозування реагентів.

INTRODUCTION

Currently, recycled water is mainly used for cooling systems in industry and energy production. However, in the future, it will also be used for municipal water supply, solving the problem of providing water to the population. An example of this is the water supply system in Johannesburg, South Africa, where wastewater and rainwater are collected after thorough treatment in aerated ponds and fed back into the water supply system.

While offering an undeniable advantage over flow-through systems in terms of water savings, recirculating systems (RS) concentrate hardness salts due to evaporation, which leads to their deposition on pipelines, fittings, and technological equipment, in particular heat exchange equipment. This results in the failure of communication and technological systems, increased costs for repair and replacement of equipment, reduced reliability, quality, and increased product cost, and ultimately, loss of competitiveness.

FORMULATION OF THE PROBLEM

It is known that the main component of deposits is calcium carbonate [1, p. 384; 2, p. 42], therefore, the work considers the hydrocarbonate subsystem of the aqueous medium. To develop and optimize measures to prevent deposits, a physicochemical model of the quality of circulating water (CW) is needed that adequately reflects the main processes occurring in CW. Below is a model based on:

1. Water-salt balance of the CW, taking into account changes in its volume, see Fig. 1; q_f, q_{CO_2}, q_{ev}, q_d, Q_S, q_{bd}, q_{fi} – feed flow, CO_2 carryover flow; evaporation, drip loss, circulation flow; blowdown, and filtration flows.

2.Kinetic equations of parallel-sequential reactions reflecting transformations in the hydrocarbonate system (HCS) [3, p.196; 4, p. 23]:

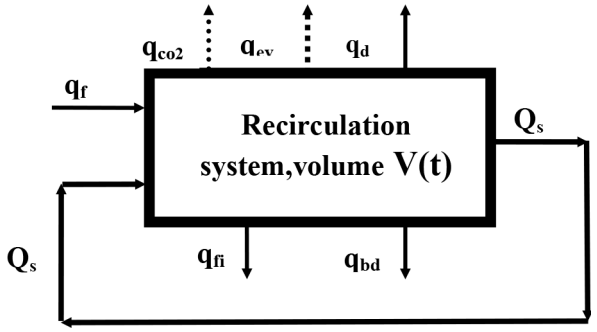

Fig. 1. Flow diagram in a recirculation system

$$CO_2 + HOH \rightarrow H^+ + HCO_3^-$$
$$HCO_3^- \rightarrow H^+ + CO_3^{2-} \quad (1)$$
$$CO_3^{2-} + HOH \rightarrow HCO_3^- + OH^-$$
$$HCO_3^- \rightarrow OH^- + CO_2$$
$$CO_{2\,water} \rightarrow CO_{2\,atm}$$
$$Ca^{2+} + CO_3^{2-} \rightarrow CaCO_{3\,kr}$$

3. Assumptions about the form of kinetic equations describing the crystallization of calcium carbonates, as well as the exchange of CO_2 between the atmosphere and the aquatic environment.

4. The law of conservation of the total amount of carbonates related to (1).

5. The condition of electrical neutrality of the aquatic environment.

6. The significant difference in the rates of reactions (1) relative to each other and the water turnover time in the system.

Further dividing the components of circulating water into two types [2, p. 40]:

– tracers – components that do not react in water with others and are not removed from the aquatic environment by gas exchange or crystallization – mainly anions of strong acids or alkaline cations;

– scale formers – components that react in an aqueous environment, precipitate, or convert to gas with subsequent evacuation – anions of weak acids or cations of alkaline earth metals, in particular hardness salts.

CALCULATION METHOD

Let us denote the molar concentration of the former by T and the latter by C. It can be shown, see, for example, [2, p. 45; 5, p.112], that the concentration of tracers satisfies the equation:

$$\frac{dT}{dt} = \frac{q_f}{V(t)}\left[T_0(t) - \frac{q_f}{q_{bd}}T(t)\right]. \quad (2)$$

$T_0(t)$, $T(t)$ are the molar concentrations of tracers in the make-up and return water at time t; $V(t)$ is the instantaneous volume of water in the return system. However, [6, p.40; 7, p. 44; 8, p. 123; 9, p. 260] did not take into account the possible change in the water volume of the recirculation system, whereas here it is assumed that there are no restrictions on changes in volume and water flows, therefore:

$$V(t) = \int_0^t \left\{q_f(t_1) - \left[q_{ev}(t_1) + q_{bd}(t_1) + q_d(t_1) + q_{fi}(t_1)\right]\right\} dt_1, \quad (3)$$

Taking into account (3), equation (2) takes the form [2, p. 42]:

$$\frac{dT}{dt} = \frac{q_f}{V(t)}\left[T_0(t) - \frac{1}{\varphi}T(t)\right], \quad \varphi(t) = \frac{q_f(t)}{q_f(t) - q_{bd}(t)}; \quad (2a)$$

φ is a dynamic factor [2, p. 42; 5, p. 112] which, when the state of the water in the treatment plant approaches a steady state or equilibrium, tends to the salt concentration coefficient: $\varphi \rightarrow k_{CL} = C_{Cl}/C_{Cl0}$ (C_{Cl0}, C_{Cl} are the concentrations of chlorides in the make-up and return water) or to the evaporation coefficient $k_{ev} = q_f/q_{bd}$, [4, p. 23], see also (2a).

The kinetic equation of the components of the second type must include terms describing their transformations into CW. For example, for the hydrocarbonate subsystem this will be the block of equations (1). Based on the kinetic equations describing reactions (1), it can be shown [1, p. 384; 3, p. 196; 5, p. 112] that the equation for the sum of carbonates

$$S_C = \left[CO_2\right] + \left[HCO_3^-\right] + \left[CO_3^{2-}\right], \quad (4)$$

is determined by the gas exchange of CO_2 with the atmosphere and the crystallization of $CaCO_3$. Representing the gas exchange process by the expression [3, p. 196]

$$-a_2([CO_2] - [CO_2]_e);$$

a_2 is the kinetic coefficient; $[CO_2]_e$ is the concentration of CO_2 in water, equilibrium with the atmosphere, and the crystallization rate is through the derivative $\frac{d[Ca^{2+}]}{dt}$, we find the kinetic equation for the sum of carbonates:

$$\frac{dS_C}{dt} = -a_2([CO_2] - [CO_2]_e) + \frac{d[Ca^{2+}]}{dt}, \quad (5)$$

refers to a closed volume. For an open volume, water exchange flows should be taken into account, which leads to the expression,

$$\frac{dS_C}{dt} = \frac{q_f}{V(t)}\left[S_C^0(t) - \frac{1}{\varphi(t)}S_C(t)\right] + \frac{d[Ca^{2+}]}{dt} - a_2([CO_2] - [CO_2]_e). \quad (6)$$

Let us use the following notation:

$$x_1 = [HCO_3^-]; \quad x_2 = [CO_2]; \quad x_3 = [CO_3^{2-}]; \quad x_4 = [H^+];$$
$$x_5 = [OH^-]; \quad x_7 = [Ca^{2+}]; \quad x_8 = [CaCO_3];$$

$[x_i]$ – molar concentration of the i-th component.

Reactions (1), considering that the water turnover time in the circulation system $T_S = V/Q_S$ is significantly longer than the characteristic times of these reactions, will be considered equilibrium reactions. Therefore, the components of the hydrocarbonate system (HCS) satisfy the following equations [3, p. 196]:

$$K_1 x_2(t) = f_1^2 x_1(t) x_4(t);$$
$$\frac{dx_7}{dt} = a_{737}[L_{37}(t) - x_3(t) x_7(t) f_2^2];$$
$$K_2 x_1(t) = f_2 x_3(t) x_4(t); \quad f_1^2 x_4(t) x_5(t) = K_w; \qquad (7)$$
$$x_1(t) + 2x_3(t) + x_5(t) - x_4(t) - 2x_7(t) = \sum K - \sum A;$$
$$D = \sum K - \sum A.$$

In equations (6) and (7), f_1 and f_2 are the activity coefficients of single- and double-charged ions; a_{737} is the kinetic coefficient; L_{37}(t) is the solubility product of $CaCO_3$ corresponding to the maximum of the $CaCO_3$ dispersion distribution function in return water; K_1 and K_2 are the thermodynamic constants of equilibrium ionization of H_2CO_3 in the first and second stages; $D = \sum K - \sum A$ is the difference in molar concentrations of cations and anions not defined by equations (7).

The system of equations (6), (7), provided that its parameters K_1, K_2, K_W, a_2, a_{737} are known, is completely defined and can be used to calculate the components of HCS in CW, including for the analysis and prediction of its scale-forming and corrosive properties or the development of measures to prevent deposits.

APPLICATION EXAMPLE

Example 1. Calculation of the rate of $CaCO_3$ release in a cooling system with cooling towers operating in steady state.

In this case: $V = \text{Const}$, $q_f = q_{ev} + q_d + q_{bd} + q_{fi}$, and CO_2 degassing occurs in the cooling tower during $\Delta T \ll T_S$ – the time it takes for water to circulate in the circulation system. For steady state,

$$\frac{dT}{dt} = 0; \quad \frac{dS_C}{dt} = 0; \quad \frac{dx_7}{dt} = -\frac{dx_8}{dt} = -\frac{d[\mathrm{CaCO_3}]}{dt}. \qquad (8)$$

From here, guided by (6), we can express the rate of $CaCO_3$ release as follows:

$$\frac{d[\mathrm{CaCO_3}]}{dt} = \frac{q_f}{V}\left(S_C^0 - \frac{1}{\varphi} S_C\right) - a_2(x_2 - x_{2e}). \qquad (9)$$

Averaging the member $a_2(x_2 - x_{2e})$ – during the water turnover in the T_S system and taking into account that for $6 < \mathrm{pH} < 10$ $S_C \approx [\mathrm{HCO_3^-}]$, we obtain:

$$\frac{d[\mathrm{CaCO_3}]}{dt} = \frac{P_f}{T_S}\left[\begin{array}{c}[\mathrm{HCO_3^-}]_0 - \frac{1}{\varphi}[\mathrm{HCO_3^-}]_S + \\ + \frac{[\mathrm{HCO_3^-}]_0 [\mathrm{H^+}]_0 f_{1,0}^2}{K_1}\end{array}\right], \quad P_f = \frac{q_f}{Q_S}. \qquad (10)$$

Thus, the rate of $CaCO_3$ release in steady-state operation of the SCV is determined by the following components:

– change in the concentration of hydrocarbonates in the CW:

$$[\mathrm{HCO_3^-}]_0 - \frac{1}{\varphi}[\mathrm{HCO_3^-}]_S;$$

– removal of CO_2 in the cooling tower: $\frac{[\mathrm{HCO_3^-}]_0 [\mathrm{H^+}] f_{1.0}^2}{K_1}$.

From equation (10), we obtain that in the absence of changes in $[\mathrm{HCO_3^-}]$ in the cooling water, calcium deposits may form due to CO_2 blowdown in cooling towers. It should be noted that in the works known to us on the kinetics of cooling water quality, see, for example, [6-10], this fact was not taken into account. If measures are taken in the circulating system to stabilize the water by acidification, for example, with sulfuric acid by dosing it into the feed, then expression (10) takes the form

$$\frac{d[\mathrm{CaCO_3}]}{dt} = \frac{P_f}{T_S} \cdot \left[\begin{array}{c}[\mathrm{HCO_3^-}]_0 - \frac{1}{\varphi}\left([\mathrm{HCO_3^-}]_S + 2[\mathrm{H_2SO_4}]_S\right) + \\ + \frac{[\mathrm{HCO_3^-}]_0 \cdot [\mathrm{H^+}]_0 \cdot f_{1,0}^2}{K_1}\end{array}\right], \qquad (11)$$

$$[\mathrm{H_2SO_4}]_S = \varphi[\mathrm{H_2SO_4}]_0.$$

Naturally, in this case, the rate of $CaCO_3$ release decreases and, at a concentration of acid in the system

$$[\mathrm{H_2SO_4}]_S = 0.5\left(\left[\varphi\left[\mathrm{HCO_3^-}\right]_0 - [\mathrm{HCO_3^-}]_S\right), \qquad (12)$$

it becomes minimal.

Note that equation (11) allows us to calculate the rate of $CaCO_3$ release by measuring the parameters of the solution given in (11) and, using the approximation $[\mathrm{HCO_3^-}] \approx L$ – total alkalinity of water, determine the concentration of sulfates that ensures an acceptable rate of $CaCO_3$ release, i.e., it can serve as a basis for automating the process of correcting the stability of the CW and thus avoiding excessive pollution of the external environment with acid discharges.

Example 2. Conditions for minimizing deposits in a recirculating system.

These conditions can be formulated as follows:

1. No $CaCO_3$ precipitation.
2. Condition of carbonate sum stability.

$$\frac{d[\mathrm{CaCO_3}]}{dt} = 0; \quad \frac{dS_C}{dt} = 0; \qquad (13)$$

where both conditions must be satisfied simultaneously.

According to (7), equations (13) are equivalent to:

$$\left.\begin{aligned} L_m(t) - x_3(t)\cdot x_7(t)\cdot f_2^2 = 0;\\ S_C^0 - \frac{1}{\varphi}\cdot S_C - a_2\cdot(x_2 - x_{2e}) = 0; \end{aligned}\right\} \quad (14)$$

Using approximation (7) for x_3(t), i.e.

$$x_3(t) = \frac{K_2 x_1(t)}{f_2 x_4(t)}$$

and for the second equation (14), taking (10), we obtain for 6 < pH < 10

$$\frac{L_{37}(t)}{f_2 K_2} - \frac{x_1(t)}{x_4(t)} x_7(t) = 0;$$

$$x_{1,0} - \frac{1}{\varphi}(x_1 + 2x_9) - \frac{x_{1,0} x_{4,0} f_{1,0}^2}{K_1} = 0; \quad (15)$$

$$x_1 + 2x_3 + x_5 = x_4 + 2x_7 - 2x_9 + D, \quad D = \sum K - \sum A;$$

$$x_1, x_7, x_9 \gg x_3, x_5.\ x_9 = [H_2SO_4];$$

D – the difference between the concentrations of cations and anions of organic matter, which are clearly not taken into account in the electroneutrality equation.

Expressing x_7 from the electroneutrality equation and substituting it into the first of (15), we obtain the value $x_{1,m}$ that minimizes the rate of carbonate release in the system. Its approximate value:

$$x_{1,m} = \frac{L_{37} x_4}{f_2 K_2 (x_9 - 0.5D)}. \quad (16)$$

Accordingly, the permissible concentration of hydrocarbonate ions in the feed water is given by the expression:

$$x_{1m}^0 = \frac{2x_9 + x_{1m}}{\varphi\left(1 + \frac{x_{4,0} f_{1,0}^2}{K_1}\right)}. \quad (17)$$

The value determines the maximum concentration of hydrocarbonates (M/dm^3) in the feed water, ensuring minimization of the rate of $CaCO_3$ precipitation in water at a given value of sulfate ion concentration x_9.

CONCLUSIONS

1. Kinetic equations describing the evolution of tracer and scale-forming components in circulating water are proposed. From the known kinetic equations for tracers [2, p. 42; 5, p. 112], expression (2) differs in that it takes into account the flows of salts removed from the system, for example, filtration, and the arbitrary dependence of the flows and volume of circulating water on time.

2. The equation for scale formers (6) is obtained based on the consideration of chemical transformations in the CW, including CO_2 degassing and $CaCO_3$ precipitation, and has no analogue in the literature known to us. This equation naturally reflects measures to stabilize the circulating water; compare expressions (10) and (11).

3. The conditions for steady-state "scale-free" operation of the circulating system are formulated in the form of (13) and (15), allowing the calculation of the limiting values of the concentrations of the main scale formers in both the circulating water and the feed water. Indeed, from (16), (17) and the last equation (15) we obtain:

$$Ca^{2+} = 0.5x_{1,m} + x_9 - 0.5D; \quad (18)$$

$$[Ca^{2+}]_0 = 0.5x_{1,m}^0 + x_{9,0} - 0.5D_0. \quad (19)$$

The values and corresponding to the steady-state scale-free operation of the recirculation system are found from formulas (16) and (17).

4. Taking into account (16)–(19), it is possible to calculate the regime of measures ensuring the stabilization of circulating water, including liming of the feedwater to achieve acceptable $[HCO_3^-]_{0,m}$ and $[Ca^{2+}]_{0,m}$.

Using (17), we will determine the portion of the makeup water that needs to be limed to stabilize the circulating water. Assuming a residual carbonate alkalinity after liming of 0.8 meq/dm^3, we find that the portion of makeup water q_{lm} for liming is determined by the ratio:

$$q_{lm} \geq q_f \frac{x_1^0 - x_{1,m}^0}{x_1^0 - 0.8}. \quad (20)$$

From (20) it follows that in this case it is necessary to lime all the feed water or use additional stabilization of the return water with inhibitors.

5. The model considered does not take into account complexes formed by Ca^{2+} ions, for example,

$$CaOH^+,\ CaHCO_3^+ \text{ and } Ca(HCO_3)_{20}.$$

As shown by calculations [10, p.16], this can lead to errors of up to 10–20 % in determining the values of $[HCO_3^-]_{0,m}$ or $[HCO_3^-]$ and, accordingly, the dose of acid stabilizing the circulating water. The above phenomena can be taken into account by appropriately modifying equation (14).

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