



Inhibition of calcium carbonate scaling by precipitation using secondary nucleation coupled to degassing with atmospheric air



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ABSTRACT

Scaling by calcium carbonate precipitation is an important challenge during exploitation of geothermal water or geothermal district heating systems. Water decarbonation is a promising solution to mitigate calcium carbonate precipitation and reduce the scaling risks. In the current work, the decarbonation of geothermal water by coupling atmospheric air bubbling with seeding was investigated. For that, several operating conditions were studied and optimized such as air and water flows, hydrodynamics, residence time, and bubbling rate. Synthetic and real solutions were tested. To reveal the efficiency of the proposed process, the addition of chemicals was avoided. Results showed that, using continuously operating reactor and under optimized operating conditions, the decarbonation rate of a calcium carbonate solution tested (4 mmol/L of CaCO_3 , $T = 50^\circ\text{C}$) can exceed 30% without using chemicals. It was also shown that the residence time in the reactor is relatively short (2.5 min) and the protection of the walls from precipitate adherence was guaranteed.

1. Introduction

The hydrological system of southern Tunisia is mainly based on the waters of underground aquifers, which currently constitute the major exploitable resources [1]. However, the use of these waters has caused scaling problems [2]. The formation of undesirable deposition, especially calcium carbonate precipitation, is mainly influenced by the high temperature and CO_2 escapement in atmospheric air which is inevitable during cooling process. Physico-chemical studies have shown that these waters are weakly supersaturated with respect to calcium carbonate [3,13]. Consequently, spontaneous nucleation in this type of solution is impossible. To mitigate scaling problems, the traditional solution is to decarbonate water with lime. But, besides the large and expensive consumption of a reagent that is not readily available in southern Tunisia, this solution generates large quantities of carbonated sludge. Thus, its implementation requires a complex apparatus using a fluidized bed crystallization support [5]. However, this process is compromised with the presence of large quantities of magnesium.

Since 1995, Tunisian geothermal water has been used in reverse osmosis process for drinkable water production [4,6,13]. In this process, the conversion rate is the most important factor [6]. The conversion rate depends on the salinity and solubility limits of salts that are likely to precipitate. The decarbonation of groundwater would have the advantage of avoiding scale deposits in the cooling towers and in the

desalination plant's sand filters, reducing the hardness of the water and conversion rate of the desalination unit. In previous work [7–9,12,13], it has been shown that seeding with aragonite can cause precipitation of calcium carbonate in calco-carbonic solutions ($\text{CaCO}_3\text{--CO}_2\text{--H}_2\text{O}$) weakly supersaturated [13,16]. In this work, a process to avoid precipitation of calcium carbonate during desalination process of ground water is proposed. In fact, the suggested process is based on decarbonation of the ground water by coupling seeding by aragonite crystals to water degassing with atmospheric air. Thus, the proposed process was tested using a laboratory reactor that operates continuously. The experimental conditions are very close to reality, since the water refrigerant uses atmospheric air, which facilitates the adaptation to industrial scale. In the first part, the technique of degassing, used with synthetic and real waters to study the different parameters that can influence the profitability of this type of decarbonation, was tested. In a second part, decarbonation of calco-carbon water is tested with a process designed to operate continuously.

2. Theory

The calco-carbonic system is considered as a multiphase system involving three simultaneous phases that imply the existence of two interfaces where the main transfer resistances are located. These resistances control the kinetics of global evolution of the system:

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Nomenclature*List of symbols*

Alc	Alkalinity (g/L)
CTE	CO ₂ transfer efficiency (–)
IAP	Ionic activity product (–)
K _s	Solubility constant of calcium carbonate (–)
K _w	Constant of the water dissociation (–)
K ₁	Constant of the first dissociation of carbonic acid (–)
K ₂	Constant of the second dissociation of carbonic acid (–)
TCa	Total calcium concentration

Q _w	Water flow (m ³ /s)
Q _{air}	Air flow (m ³ /s)
V _w	Volume of water (L)
V _g	Volume of air (L)
Ω _{/gypsum}	Super-saturation of the solution regarding the gypsum (–)
Ω _{/CaCO₃}	Super-saturation of the solution regarding the calcium carbonate (–)
Ω _{Aragonite}	Super-saturation of the solution regarding the aragonite (–)
Ω _{Calcite}	Super-saturation of the solution regarding the calcite (–)
Ω _{/CCM}	Super-saturation of the solution regarding the calcium carbonate monohydrated (–)

2.1. Gas/liquid interface

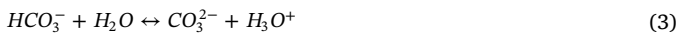
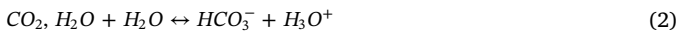
We speak of equilibrium of adsorption-desorption of CO₂ which will be described by Henry's law following this equilibrium relation [10]:

$$P_{CO_2} = K_H \cdot (CO_2) \quad (1)$$

K_H is Henry's coefficient

2.2. Liquid phase

A part of dissolved CO₂ will be hydrated and then ionized, giving rise to the HCO₃[–] and CO₃^{2–} ions which will be characterized by their activities (HCO₃[–]) and (CO₃^{2–}). The dissociation reactions are [11,12]:



The application of the law of mass action to ionization reactions yields [11]:

$$K_1 = \frac{(HCO_3^-)(H_3O^+)}{(CO_2)} \quad (4)$$

$$K_2 = \frac{(CO_3^{2-})(H_3O^+)}{(HCO_3^-)} \quad (5)$$

With K₁ and K₂ are respectively the constants of the 1st and 2nd dissociation of the carboxylic acid. These ionizations release H₃O⁺ and OH[–] whose activities are connected by the ionization constant of water:

$$K_w = [H^+] \times [OH^-] \quad (6)$$

The ionic concentrations must respect the electric neutrality condition of the solution expressed by the following relation:

$$2[Ca^{2+}] + 2[H_3O^+] = [OH^-] + 2[CO_3^{2-}] + [HCO_3^-] \quad (7)$$

The partial pressure of CO₂ at the gas/liquid interface is a main parameter in the precipitation process of calcium carbonate [13,16,17]. The degassing of a calco-carbonic solution by atmospheric air reduces the partial pressure of CO₂. Eqs. (1)–(4) can be combined to calculate this parameter:

$$P_{CO_2} = K_H (CO_2) = K_H [HCO_3^-] \gamma_{HCO_3^-} 10^{pK_1 - pH} \quad (8)$$

Eq. (8) clearly shows that when the partial pressure of CO₂ decreases the pH increases.

2.3. Solid/liquid interface

Three compounds are likely to precipitate. Carbonate, hydrogen carbonate and calcium hydroxide. Since the calcium carbonate is the least soluble among the compounds mentioned above, its solubility product will be considered:

$$(Ca^{2+}) \times (CO_3^{2-}) = K_s \quad (9)$$

The super-saturation depends on both molar concentration of the constituent ions and their degree of ionization, which itself depends on several parameters such as pH, temperature and ionic strength [14].

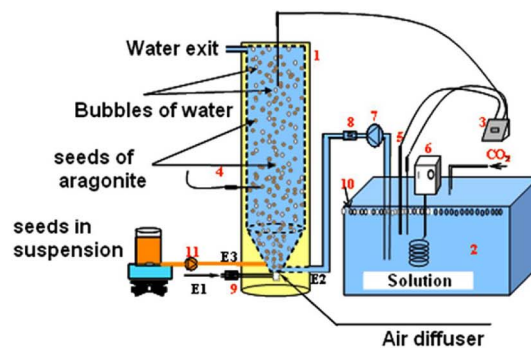
$$\Omega = \frac{[Ca^{2+}][CO_3^{2-}]}{K_s} \quad (10)$$

with

$$(Ca^{2+}) = \gamma_{Ca^{2+}} [Ca^{2+}] \quad (11)$$

Eq. (9) can be written as:

$$(CO_3^{2-}) = (HCO_3^-) 10^{pH - pK_2} = \gamma_{HCO_3^-} [HCO_3^-] 10^{pH - pK_2} \quad (12)$$



1: Plexiglas reactor; 2: Thermostatic bath; 3: pH-meter; 4: pH electrode; 5: Temperature sensor; 6: Immersion heater; 7: Water pump; 8: Flow-meter (water); 9: Flow-meter (air); 10: Polystyrene; 11: Injections pump.

Fig. 1. Experimental device of the decarbonation technique.

1: Plexiglas reactor; 2: Thermostatic bath; 3: pH-meter; 4: pH electrode; 5: Temperature sensor; 6: Immersion heater; 7: Water pump; 8: Flow-meter (water); 9: Flow-meter (air); 10: Polystyrene; 11: Injections pump.

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