



Sand filters scaling by calcium carbonate precipitation during groundwater reverse osmosis desalination

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ABSTRACT

To face water scarcity in the south, Tunisia is relying on the use of fossil water from wells as feed water in reverse osmosis desalination. This geothermal water reaches the surface with specific characteristics ($50\text{ }^{\circ}\text{C} < T < 70\text{ }^{\circ}\text{C}$, hardness $\geq 10\text{ mM}$, $P_{\text{CO}_2} \approx 5.10^{-2}\text{ atm}$, salinity $\geq 3\text{ g/L}$). During the cooling stage, an extensive scaling takes place in the cooling towers following the release of CO_2 . This phenomenon continues even in the piping system. The scaling reached the sand filter in the pre-treatment process. In the current work, the scaling in sand filters was investigated. Firstly, the nature of scaling was identified. X-Ray Diffraction (DRX) of the sand samples revealed the existence of significant quantities of aragonite form of calcium carbonate (CaCO_3). DRX and scanning electron microscopy (SEM) analysis revealed the presence of aragonite crystals in the raw water. Investigations showed that these crystals trapped in the sand filter act as seeds inducing secondary nucleation of CaCO_3 . The study showed that the raw water is over saturated with respect to both calcite and aragonite, while it is under saturated relative to the CaCO_3 monohydrate form. It was suggested that the feed water is at meta-stable state, and crystal nucleation can only be driven through seeding. Laboratory experiments using the raw water with simulated sand filter confirmed the inevitable secondary nucleation supported by aragonite crystals. Consequently, the suggested solution was a water acidification ahead of the sand filtration operation instead of past of it in the desalination process.

1. Introduction

Water is an indispensable resource for life. This essential source is widely used in several fields such as agriculture, industry and domestic needs. In nature, there are four main sources of raw water; rainwater, surface waters, subterranean waters and seawater [1]. All these categories revealed different qualities made up of the different salts naturally dissolved [2,3]. Nowadays, the world is facing the problem of water scarcity [4,5]. To meet the increase of water demand and limited supply of drinkable water, the need for alternative fresh water sources is strongly required. Desalination is one of the most useful processes that provides alternative and sustainable solution for water scarcity problems [6,7]. Water desalination techniques, especially based on membrane separation, are in full expansion in the world. Like many Mediterranean countries, Tunisia has installed several reverse osmosis (RO) desalination plants. In literature, most studied problems in desalination process are mainly related to membrane fouling and scaling [8–12]. This is axiomatic due to the fact that the membrane is the key piece in separation process. Consequently, intense researches were

made to provide an adequate pretreatment to minimize the risk of fouling and salt depositions that reduce the lifetime and membrane performance [13–15]. However, in our best knowledge, fouling and scaling in filters are considerably neglected. Filters guarantee the well-functioning of the process. Therefore, filters performance also should be optimized. In the southern of Tunisia (RO desalination plant, Chott El-Fejjej- Gabes district), the treated raw water is essentially groundwater. Fossil water from boreholes whose depths vary between 1000 and 3000 m are exploited [16]. The water temperature at the outlet of boreholes varies between 50 and 70 °C, its calcium concentration and alkalinity are of the order of 10 and 2 mM, respectively, the salinity is $> 3\text{ g/L}$ and the dissolved carbon dioxide CO_2 is relatively high ($P_{\text{CO}_2} \approx 5.10^{-2}\text{ atm}$) [16]. Drilling rates vary from 45 to 140 L/s [17]. Chott El-Fejjej desalination plant has a series of sand filters which suffer from the increase of sand filtering mass due to the accumulation of inorganic deposit that adheres to the sand grains. This problem requires slow and expensive regeneration. During the cooling step, which is a fundamental operation before water use, a gigantic quantity of scale is deposited following the escapement of CO_2 . The deposition continues in

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the pipes and causes a severe problem to the exploitation of this water. The decarbonation of the Chott El-Fejje water would have the advantage of avoiding deposits of scale in the cooling towers and in the sand filters, reducing the hardness of the raw water and the recovery rate of the desalination plant. In this situation, the conventional solution is to decarbonate with lime. However, besides the large and expensive consumption of such reagent, which is not readily available in the Gabes desert, this process generates large amounts of carbonated sludge. Thus, if this treatment is considered, it supposes a complex apparatus using a fluidized bed crystallization support [18] whose good functioning may be compromised with the presence of large amounts of magnesium [18]. In this work, an alternative solution was suggested. The technical intervention consists of avoiding the nucleation of calcium carbonate in the raw water. For that, the raw water was, firstly, characterized with “calco-carbonic equilibrium” point of view. Secondly, the increase of sand filter mass was estimated experimentally. Thirdly, the sand filter was simulated in lab-scale for testing synthetic and real water. The filter was designed to operate continuously and experimental conditions were very close to reality, since Chott El-Fejje’s refrigerants are using air ventilation, which would facilitate the adaptation on an industrial scale. Finally, the increase of the sand filter mass was assessed to prove that the secondary nucleation of calcium carbonate is inevitable. Consequently, the most suitable solution to optimize the performance of the sand filters was taken according to experimental results.

2. Meta-stability in calco-carbonic system

In the calco-carbonic system, hydrated forms play an important role in the process of calcium carbonate nucleation [25]. In fact, spontaneous nucleation, called primary, can only take place if one of the hydrated forms, called precursor, appears. If the calco-carbonic solution is supersaturated with respect to the amorphous calcium carbonate form (ACC) (see Zone A1 – Fig. 1), ACC variety plays the role of a precursor for a homogeneous and instantaneous nucleation, regardless the nature of the walls and foreign particles [19,20]. In the case where the product of the ionic activity of the solution is between the solubility product of the ACC and that of the calcium carbonate monohydrated (CCM) (Zone A2 - Fig. 1), heterogeneous primary nucleation takes place, the precursor here is the monohydrate form. This nucleation depends strongly on the nature of the walls and foreign particles, especially for low saturation solutions with respect to the monohydrate form. If the state of super-saturation does not result in the formation of one of these precursors (CCM and ACC), the system remains in a state of meta-stability. In other words, a super-saturation of the calco-carbonic system with respect to the anhydrous forms is insufficient for a spontaneous nucleation of the CaCO_3 . The meta-stability zone of the

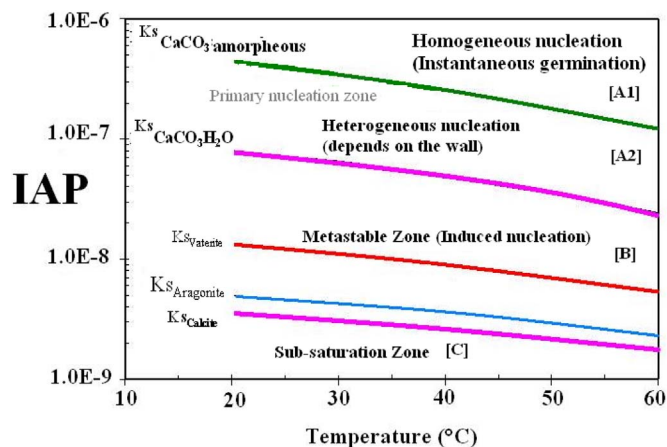


Fig. 1. Meta-stability diagram of calco-carbonic system.

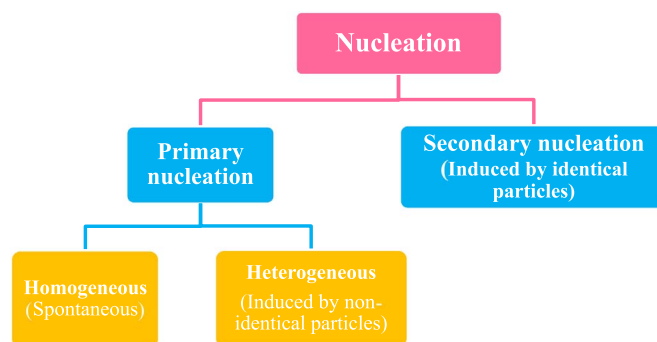


Fig. 2. Schematic of nucleation mechanisms.

calco-carbonic system has been delimited thermodynamically [21], it lies between the calcite solubility product and CCM solubility product (Zone B – Fig. 1). This meta-stability can be broken by seeding with CaCO_3 crystals. Therefore, a secondary nucleation supported by seed crystals of CaCO_3 occurs.

2.1. Mechanism of nucleation

Nucleation is a process in which the free or dissolved ions or molecules gather and organize to form the so-called germ. The appearance of the crystals can only be achieved when a degree of super-saturation is reached. Three stages can be distinguished during the precipitation of calcium carbonate: nucleation, dehydration and crystal growth. The process begins with the ions that agglomerate forming a “cluster”. The grouping of these aggregates forms a colloidal nucleus which grows and gives a stable crystal. Primary nucleation and secondary nucleation are distinguished (Fig. 2). A) Homogeneous nucleation: It is obtained when the nucleus develops only in the liquid phase and is not influenced by the presence of impurities in the reaction medium. The number of molecules in a stable germ can vary from about 100 to several thousand. The actual formation of such a nucleus results from the simultaneous collision of the number of necessary molecules. This is a highly unlikely phenomenon. The process of forming such a germ involves a certain energy called the free energy of activation of nucleation. B) Heterogeneous nucleation: Nucleation is said to be heterogeneous when the nucleus develops on a support or in the presence of impurities. These impurities can act as an accelerator as well as an inhibitor [24]. It is generally appropriate that heterogeneous nucleation can be initiated to a lower degree of super-saturation than homogeneous nucleation. The variation in free enthalpy associated with heterogeneous nucleation is lower than that associated with homogeneous nucleation [21,24]. The secondary nucleation, in a weakly supersaturated solution, is induced and supported by seed crystals of the same nature as the precipitate. In this type of nucleation, the affinity between the crystals of the solute formed and the seed crystals (solid surface) is total, which corresponds to a critical energy of zero nucleation [21,24].

3. Materials and methods

3.1. Sand filter description and problematic

The separation is carried out by means of a static filter bed. The filter should be made by chemically insoluble material which is inert with respect to aggressive water and capable of undergoing the intense friction caused by the sand washes without crumbling. A conventional filter is presented in Fig. 3. It consists of: *i*) a filter bed made of sand and anthracite, *ii*) a gravel support, *iii*) a filter background, *vi*) a device for washing the sand bed, *v*) a washing water discharge chutes, and *iv*) valves for water inlet and outlet as well as control devices for the various phases of the operation.

The filter feed is provided by overflowing the water from the middle

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