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The influence of magnetic water treatment on $CaCO_3$ scale formation in membrane distillation process

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ABSTRACT

In the present study the effect of using magnetic water treatment in order to reduce carbonate deposit during membrane distillation was investigated. Tap water containing 2.16 mmol HCO_3^- dm^{-3}, which was additionally enriched with bicarbonates (alkalinity 2.72 mmol HCO_3^- dm^{-3} and 3.61 mmol HCO_3^- dm^{-3}) was used in the study. To generate a magnetic field a commercial device, called the magnetizer, was used. The magnetic field originated from two S–S type permanent magnets, 0.1 T each. For comparison purposes the membrane distillation with/without a magnetic pre-treatment of the feed were conducted. The distillation process proceeded at 358 K. When the feed was warmed up, large amounts of $CaCO_3$ deposit were formed inside the experimental installation. SEM–EDS and XRD were used to investigate the chemical composition and morphology of the precipitate. The precipitate was also forming on the membrane surfaces, which resulted in a decrease of modules efficiency. It was found that as a result of magnetic pre-treatment larger crystallites were formed and the deposit was more porous. For this reason a decrease in the permeate flux was significantly smaller when the magnetizer was used. The $CaCO_3$ deposit was found to be mostly in the form of calcite in each investigated case.

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1. Introduction

In membrane distillation (MD) water evaporates from a hot feed through the pores of non-wetted membranes. The membranes retain salts and other non-volatile solutes, which enable the production of high purity water [1–4]. However, MD process is hindered by the presence of bicarbonates dissolved in raw water, which undergo the decomposition during the feed heating followed by the precipitation of carbonates on the membrane surface [1,5]. Scaling causes a progressive wettability of the membrane, and as a result a decline of both the permeate flux and separation efficiency was observed [1,2,6].

The operating conditions of MD process influenced on the scaling intensity, and it was indicated that the formation of $CaCO_3$ deposit could be hindered using a feed temperature below 343 K and the feed flow rate above 0.5 m s^{-1} [1,7]. Moreover, HCO_3^- ion concentration can be reduced at the pretreatment stage, e.g. by chemical water softening or by using pressure-driven membrane processes [2,8,9]. A complete removal of bicarbonates from feed water is an effective solution and can be achieved by acidifying the feed to pH 4, which is associated with the use of large amounts of acid [2].

The author's previous studies seem to suggest that only by using the water pretreatment it is probably not possible to eliminate scaling in MD process [2,5–8]. Moreover, as a result of removing the majority of HCO_3^- ion from water, the carbonates instead of crystalline deposit formed an amorphous precipitate with increased amount of silicon. This kind of amorphous precipitate accelerated a decrease in MD efficiency [5].

The formation of $CaCO_3$ crystals have a tendency to undergo a selective adsorption of several ionic species, which in the deposit being created enable the formation of other compounds leading to membrane scaling, e.g. $CaSO_4$ [10]. Thus, membrane scaling can be limited by the separation of $CaCO_3$ deposit in a pre-filtration element assembled at the inlet of the MD module. The efficiency of heterogeneous crystallization on net-filter surfaces decreased when the deposit layer covered the entire surface of pre-filter element. Therefore, the formed deposit should be systematically removed from this pre-filter, e.g. by rinsing it with acid solutions. A periodical rinsing of filter nets by HCl solutions did not have a negative influence on the membrane performance, and the MD module efficiency was stabilized during the long-term investigations [11].

The efficiency of pre-filter elements depends on the rate of nucleation/crystallization and on the size of forming crystals. Several previous papers suggest that a water flow through a magnetic field can significantly affect these parameters [12–20]. Magnetic water treatment (MWT) leads to the formation of $CaCO_3$ particles in the bulk of scaling water, which cannot precipitate on the walls

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of distributions pipes and other pieces of equipment. These particles are carried away by the water flow and can be eliminated by removing or filtering the resulting calcareous mud [12–16]. For this reason MWT is becoming an alternative to the chemical treatment in preventing scale formation in the industrial and some other commercial water systems [17–19]. Various magnetic, electromagnetic and electrostatic devices offered to control lime scale formation are sold worldwide for domestic and industrial applications. Typical products incorporate arrangements of permanent magnets [15,17,18].

Unlike chemical water softening, a magnetic treatment should have no direct effect on water chemistry. It is claimed to alter the morphology and adhesion of calcium carbonate scale [13,18–20]. $CaCO_3$ has three anhydrous crystalline polymorphs: vaterite, aragonite, and calcite. Additionally, three metastable forms: amorphous calcium carbonate (ACC), crystalline monohydrate ($CaCO_3 \cdot H_2O$), and hexahydrate ($CaCO_3 \cdot 6H_2O$) have also been reported [21]. The particles of calcium carbonate that were crystallized in a magnetic field consisted mainly of a mixture of aragonite and vaterite, however, significant amounts of calcite were also observed [13–16]. Calcite is usually associated with hard scale whereas aragonite and vaterite give rise to a softer type of scale that is easily removed [19,22]. According to several authors MWT would tend to reduce the nucleation rate and to accelerate a crystal growth [12,20,23].

The magnetic effect has been attributed to a number of potential mechanisms broadly divided into physical and chemical ones. The chemical mechanisms include a modification of crystal liquid interface including the changes in crystallite nucleation [14]. Several authors claim that the important factors which promote magneto–hydrodynamic forces (responsible for the changes in crystallization) are conductivity of solution, linear velocity of fluid, and flux density of magnetic field [12,13,15,19,23]. It was found that even a weak magnetic field ($B = 0.1$ T) influenced aragonite/calcite ratio in precipitated $CaCO_3$ in ground water [17].

A significant influence on the crystalline polymorphs of $CaCO_3$ has the flow turbulence and density of magnetic field. For weak magnetic field (0.16 T) and a flow character close to a laminar region (Re = 1900–3000) the calcite was formed as a result of MWT using [23]. In case of the laminar flow, but stronger magnetic field (0.5–1.3 T) the application of MWT promotes the precipitation of aragonite polymorph of $CaCO_3$, however, calcite also was indicted [19,22]. Using the turbulent flow (Re = 6000–7000) and strong magnetic field (0.4–1.5 T) aragonite was mainly formed [20,22].

Owing to the complexity of the investigated mechanisms MWT effects are variable and depend to a large extend on the process conditions. The results of experiments are affected even by the way in which working solutions are prepared. Two methods of $CaCO_3$ precipitation are commonly used. In the first method, sample solutions of calcium hydrogen carbonate were prepared by dissolving finely ground calcium carbonate powder of analytical purity in deionised water and bubbling the suspension with CO_2. As carbon dioxide is removed from the system by bubbling air through the solution, $CaCO_3$ particles are precipitated [12,19,22,23]. In the latter method, crystals are formed by mixing magnetically pretreated solutions of $CaCl_2$ and Na_2CO_3 [16,17]. The results showed that MWT of Na_2CO_3 solutions favored the formation of aragonite polymorph of $CaCO_3$, while magnetic exposure of $CaCl_2$ solutions caused no noticeable effects [22]. Moreover, it was revealed that different impure elements (e.g. Fe^{3+}, Zn^{2+}, Mg^{2+} and Cu^{2+} ions) have a significant influence on the crystallization form of $CaCO_3$ [15,19].

The efficiency of magnetic treatment depends on the material used to build the pipe from which water flows through a magnetic gap of the device. The presented results demonstrate that the magnetic devices work also with non-conducting pipes or bodies [15,16,23,24]. However, the concentration of ionic calcium was reduced by 18% for a pipe made of PVC, whereas it was reduced by 28% for stainless steel and copper [15]. It is important to be able to use plastics, because the products of metal corrosion fouled the MD modules [25].

MWT in the membrane processes was used to pretreat water feeding reverse osmosis [16]. In this case, the magnetic treatment did not reduce the rate of membrane scale formation, but the deposit forming from magnetically treated water tended to be more granular and much more substantial. The magnetic treatment increased the total amount of precipitate, which was formed in large crystals, which made it easier to separate them in the filters.

Effects of magnetic water treatments on the physical parameters, such as scale deposition, conductivity and pH, are reported not only in the scientific papers, but also in the industrial technical reports [26,27]. A commercially available magnetic treatment device was used for several applications. The Magnetizer Group Inc. (USA) has sold a wide range of products based on ceramic–magnet-array technologies to provide effective energy reduction performance for commercial and industrial use, e.g. for the prevention of scale and corrosion or for the reduction of algae and biocide additives [26]. MWT water treatment was also used for the protection of heat exchangers of 1 GW electric power plant in Łaziska (Poland) [27]. After several months of installation exploitation only a small amounts of soft, amorphous deposits, mainly silica hydrosol, was detected. In work [3] the authors successfully applied MWT for water feeding a large apartment block. The results were comparably good as those for the chemical softening of water.

A review of the literature has revealed that most of reported successful applications have occurred in continuously recirculation systems enabling repeated treatment of the process water. A similar situation is to be found in MD process, where the heating feed is repeatedly recirculated through MD modules. The aim of the study was to examine whether or not magnetic field can lower scaling in MD process.

2. Experimental

The investigations were carried out using the experimental set-up shown in Fig. 1. The feed and distillate streams flowed co-currently from the bottom to the upper part of the vertically positioned capillary MD module (0.0127 m^2). For each experimental series a new module (MK1–MK4) was used. Nine hydrophobic polypropylene membranes (Accurel S6/2 PP, Membrana, Germany), with the outside/inside diameter equal to d_{out}/d_{in} = 2.6 mm/1.8 mm, were assembled inside these modules (effective length 0.25 m). The applied membranes have the pore sizes with the nominal diameter of 0.22 μm, and the porosity of 73% (the manufacturer's data). During all the experiments the feed was supplied inside the membrane capillaries (0.58 m/s – Reynolds number Re = 3020), whereas the distillate flowed (0.116 m/s) on the shell-side of MD module. The inlet temperatures of distillate (293 K) and the feed (358 K) were constant for all the experiments. The temperatures were measured using thermometers with ±0.2 K accuracy.

The results presented by other authors indicated that the effect of MWT action for raw water can be different from those obtained for the analytical grade solutions containing only the following ions: Ca^{2+}, Na^+, Cl^- and HCO_3^-. Therefore, to obtain the results of MWT application in the MD process close to those obtained during the desalination of natural surface water, the feed prepared based on the tap water was used in the MD investigations. This tap water was produced from surface water collected from a lake, which is subjected to chlorination after the treatment. The concentration

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