

Deacidification through calcium carbonate dosing in combination with ultrafiltration

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ABSTRACT

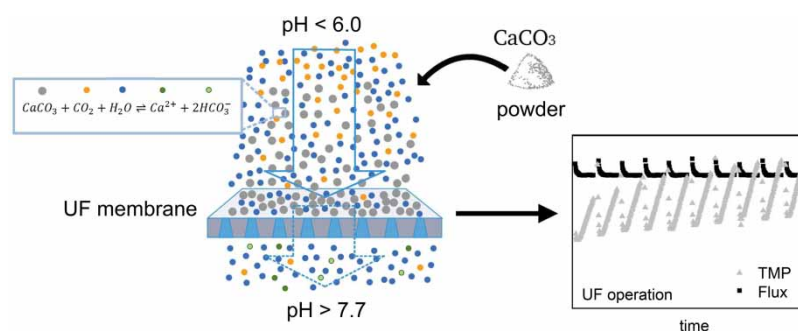
Soft acidic waters are often treated for drinking water purposes by using limestone filters to attain chemical equilibrium. The present study investigated the process parameters of a relatively new process combination in which powdered calcium carbonate (CaCO_3) was added prior to an ultrafiltration (UF). In order to reach the targeted pH value (≥ 7.8), dosing concentration, type of material and retention time were evaluated in pilot-scale experiments. The deacidification followed the same kinetics as for limestone filtration and yielded similar filtrate characteristics with dosing concentrations of 20 and 40 g/L CaCO_3 . No significant increase in transmembrane pressure was observed during the operation of a pilot-scale UF module at low flux ($34 \text{ L m}^{-2} \text{ h}^{-1}$). Critical flux was determined in a laboratory scale to evaluate the potential impact of CaCO_3 particles on the UF operation. Stepping-flux experiments revealed the presence of fouling only at high-dosing concentrations, resulting in a critical flux of $55 \text{ L m}^{-2} \text{ h}^{-1}$. At a higher flux, a CaCO_3 -fouling layer was formed, which decreased the membrane's permeability by 20% over 5 h. Considering that effective air-enhanced backwash and acidic chemical cleanings will be implemented in large-scale applications, the investigated process combination promises to be an appropriate treatment technology for turbid and soft acidic waters.

Key words: calcium carbonate, critical flux, deacidification, fouling, ultrafiltration

HIGHLIGHTS

- Dosing of powdered CaCO_3 as a deacidification alternative followed by ultrafiltration.
- Absence of CaCO_3 fouling at a low-flux UF operation with air-enhanced backwash.
- Determination of critical flux for the filtration of CaCO_3 suspensions.
- Presence of irreversible fouling at high CaCO_3 concentration and operating flux.

GRAPHICAL ABSTRACT



INTRODUCTION

Soft water, which is acidic due to excessive carbon dioxide (CO_2) content, requires treatment to avoid corrosion in the drinking water supply networks. Technical rules for the distribution of such waters have been introduced to reduce the CO_2 concentration, thereby increasing pH and approaching a chemical equilibrium state and buffering capacity. The German

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drinking water ordinance specifies a maximum calcite dissolution capacity of 5 mg/L calcium carbonate (CaCO₃), or alternatively, a minimum pH value of 7.7 (Trinkwasserverordnung – TrinkwV 2001). Therefore, reaching a pH value equal to the saturation pH or higher than 7.8 was the aim of this study.

Different mechanical and chemical deacidification processes are used in practice to deal with acidic waters. A common technique is to treat soft waters from natural sources by filtration through alkaline materials such as granular calcium carbonate (Mutschmann & Stimmelmayer 2007). This process, also known as limestone filtration, reduces the CO₂ content and at the same time increases the calcium (Ca²⁺) and bicarbonate (HCO₃⁻) concentrations following the reaction in Equation (1):



The reaction to reach the lime-carbonic acid equilibrium or the target of deacidification takes place over a period of time that is determined by the filter's dimensions and that is referred to as empty bed contact time (EBCT). The raw water characteristics (i.e., temperature, pH, acid and base capacities) determine the quantity of calcium carbonate that can be dissolved and, hence, the contact time needed for the required pH increase. However, for very acidic soft waters, long contact times are required, which may result in impractically large filter volumes (Mutschmann & Stimmelmayer 2007).

The dissolution kinetics of calcium carbonate are controlled by the particle surface area (Sjöberg 1976; Gude *et al.* 2011; Hasson *et al.* 2017). A larger available surface area increases the rate of dissolution. As a result, an alternative to calcium carbonate filtration is the addition of powdered CaCO₃ material, which has a higher specific surface area, thus enhancing its dissolution and reducing the required contact times. This method entails a dosing larger than the calcite dissolution capacity of the water to significantly speed up the reaction (Gude *et al.* 2011). A further process step, such as ultrafiltration (UF), is then required to remove the excessive CaCO₃ powder suspension.

The dissolution of granular and powdered CaCO₃ has been investigated for many years till now. The literature covers the different parameters as well as approaches to model the dissolution process (Weyl 1959; Morse 1974; Sjöberg 1976; Plummer *et al.* 1978; Dreybrodt 1981; Yamauchi *et al.* 1987; Hasson *et al.* 2017). However, no research has been done on the kinetics in a process combination of dosing powdered CaCO₃ in the UF feed.

Previous studies on membrane fouling with calcium carbonate mostly focus on its interaction with natural organic matter (Kweon & Lawler 2004; Renou *et al.* 2009). Nevertheless, CaCO₃ particles by themselves may have a substantial fouling potential through pore clogging and cake buildup that can ultimately limit the productivity of the UF membranes (Gao *et al.* 2011), leading to the need to identify sustainable operation conditions to make this process combination feasible.

One method to achieve optimal operation conditions of membrane processes, considering that fouling and cake layer formation are the major factors for their performance deterioration, is by establishing a critical flux. In dead-end UF, this concept is used to determine the threshold between low or near constant and high fouling rate regions (Bacchin *et al.* 2006; Field & Pearce 2011).

In this study, the process combination of deacidification through a dosing of calcium carbonate followed by UF was evaluated as a treatment alternative for soft acidic water. The feasibility of the overall process was first investigated at the pilot scale to determine the operational conditions for the deacidification. Additionally, the fouling potential on UF membranes of selected CaCO₃-dosing concentrations was assessed in a small laboratory-scale filtration unit by determining the fouling rate and critical flux.

MATERIALS AND METHODS

Feed water

Synthetic feed waters were prepared to simulate soft acidic spring water from the Black Forest region (Germany). The synthetic solutions consisted of mixtures of demineralized water, small amounts of tap water (Karlsruhe, Germany) and carbonated water (produced with tap water) at room temperature (18–21 °C). Table 1 lists the relevant water quality parameters used as target values to approximate the characteristics of the raw water of interest.

Pilot plant setup

The process combination of deacidification by a dosing of calcium carbonate powder and a subsequent UF system was investigated in a pilot plant. As depicted in Figure 1, experiments were carried out by pumping synthetic feed water from a feed tank at 100 L/h through an inline dosing point (powdered CaCO₃) to an intermediate tank.

Table 1 | Relevant target parameters for feed water preparation

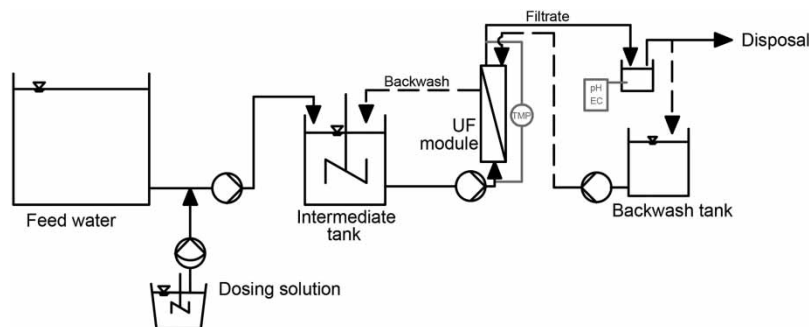
Parameter	Target value	Unit
pH	5.7–6.0	
Electrical conductivity (25 °C)	25–35	μS/cm
Calcite dissolution capacity	80–90	mg/L CaCO ₃
Base capacity, K _B 8.2	0.48–0.53	mmol/L
Acid capacity, K _S 4.3	0.15–0.17	mmol/L
Calcium	3.0–4.1	mg/L
Magnesium	0.62–0.66	mg/L
Sodium	2.8–3.3	mg/L
Potassium	1.7–2.3	mg/L

Suspensions of the powdered CaCO₃ with the desired dosing concentration were produced with feed water and continuously dosed at 2 L/h. Two CaCO₃ products, Kalksteinmehl (Rheinkalk GmbH, Germany) and Juraperle BM (Eduard Merkle GmbH, Germany), were used for the experiments. According to the manufacturers' datasheets, Kalksteinmehl (KM) has a particle size distribution of 89–99% under 125 μm and 74–84% under 63 μm, while Juraperle (JP) has 99.9% of particles under 40 μm and 94% under 8 μm. However, particle count measurements performed in the 1–100 μm size range for diluted suspensions of the materials showed no significant difference between both products. Compared with typical CaCO₃ filter material with a grain size of around 1–2 mm, the powdered material offers a significantly larger surface area and, therefore, faster dissolution kinetics are expected.

The water was mixed in the intermediate tank and then pumped at 130 L/h (34 L m⁻² h⁻¹) into a hollow-fiber UF membrane module (at an effective area of 3.8 m², polyethersulfone/polyvinylpyrrolidone blend, Aquaflex 6.2–0.8 UFC, X-Flow BV, Netherlands), operated in a dead-end inside-out mode. A pressure transducer was used to record the transmembrane pressure (TMP). During each experiment, a total of four 30-min filtration cycles were carried out. Backwash steps with UF filtrate enhanced with air on the feed side were performed for 90 s between the filtration cycles. The backwash water was returned to the intermediate tank, thus recirculating the dosed material and thereby prolonging the contact time of water with calcium carbonate.

Turbidity of the water entering the UF module was measured (portable turbidimeter, Hach Lange GmbH, Germany) once every filtration cycle. The pH and electrical conductivity of the filtrate were continuously recorded with a multiparameter meter (Multi 3620 IDS, Xylem Analytics Germany). One sample of the feed water and two of the filtrate were taken for acid and base capacity measurements and consequently to determine the HCO₃⁻ and CO₂ concentrations, as described in the DIN 38409–7 and DIN 38404-10, and for cation analysis using Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES 725, Agilent Technologies).

Throughout the experimental phase, the dosing concentration, the type of CaCO₃ powder and the volume in the intermediate tank (i.e., the system's contact time) were varied.

**Figure 1** | Pilot plant schematic.

Small laboratory-scale filtration

A small lab-scale filtration unit was used to simulate the CaCO₃ loading and, hence, its fouling potential on the UF membrane at the selected dosing concentrations and under controlled flux conditions. The highest turbidity value observed during the pilot experiments at the selected conditions was used as a reference to produce the feed suspensions to be filtered. The target feed concentration was prepared by adding the CaCO₃ material to the synthetic feed water as batch and stirred during the experiments.

Single-fiber membranes (approximate area of 30 cm², Multibore 0.9, Inge, Germany) manufactured with seven capillaries for dead-end inside-out operation were used to conduct the filtration experiments. A new membrane was used for each of the dosing concentrations.

Critical flux determination

The filtration experiments were performed to determine the threshold between fluxes producing zero or low and high fouling rates. This critical flux was derived by means of the stepping-flux method and fouling rate assessment outlined in *Le Clech et al. (2003)* and *Tuczinski (2018)*. This approach consists in performing a series of filtration cycles over a set time step (Δt). The flux was increased (ascending phase) at every cycle by a flux step (ΔJ) and subsequently decreased repeating some of the flux steps (descending phase). The flux-TMP profile was evaluated by calculating the TMP derivative, i.e., fouling rate $\Delta TMP/\Delta t$, as the difference between the final (TMP_f) and the initial (TMP_i) transmembrane pressure within each filtration step:

$$\frac{\Delta TMP}{\Delta t} = \frac{TMP_f - TMP_i}{\Delta t} \quad (2)$$

The time steps and fluxes were arbitrarily selected for the experiments and only two fluxes were repeated during the descending phase. In between filtration cycles, backwash with demineralized water was done for 90 s at $>180 \text{ L m}^{-2} \text{ h}^{-1}$. Critical flux is defined as the threshold flux from where the TMP considerably increases.

RESULTS AND DISCUSSION

Deacidification through CaCO₃ dosing

The pilot plant experiments and the variation of the operational parameters allowed the determination of the process conditions needed to reach the target water quality in terms of the pH value. *Table 2* lists the most relevant operational parameters and the analytical results of the feed and filtrate samples for some of the experiments.

Based on the established target pH of 7.8, the results showed that by operating with an intermediate tank of 75 L and a dosing concentration of 20 g/L of either KM or JP, an effective deacidification could be achieved.

Table 2 | Feed and filtrate characteristics from pilot plant experiments (18–21 °C)

Dosing	Tank volume		pH		Elec. Conductivity		CO ₂		HCO ₃ ⁻		Calcium		Magnesium	
					Feed	Filtrate	Feed	Filtrate	Feed	Filtrate	Feed	Filtrate	Feed	Filtrate
CaCO ₃	g/L	L	Feed	Filtrate	Feed	Filtrate	Feed	Filtrate	Feed	Filtrate	Feed	Filtrate	Feed	Filtrate
KM	15	75	6.0	7.7	29	116	0.57	0.05	0.20	1.07	4.0	21.1	<0.50	0.55
KM	20	75	5.9	8.1	29	122	0.54	0.01	0.20	1.12	4.0	22.2	0.50	0.57
KM	40	75	6.0	8.1	29	131	0.59	0.01	0.36	1.25	3.9	24.6	<0.50	0.60
JP	20	75	6.0	8.1	27	119	0.53	0.01	0.18	1.11	3.7	22.0	<0.50	<0.50
JP	20	30	5.9	7.8	27	131	0.54	0.04	0.19	1.20	3.7	23.4	<0.50	0.51
JP	20	30	6.0	7.9	28	131	0.52	0.02	0.19	1.20	3.8	23.8	<0.50	0.51
JP	40	30	6.0	8.0	28	136	0.56	0.02	0.19	1.30	3.9	25.8	<0.50	0.53

Considering the importance of the contact time for a complete deacidification to take place, the intermediate tank volume was further reduced from 75 to 30 L, corresponding to a shortening of the contact time from around 35 to 14 min. The experiments revealed that with this lower intermediate tank volume, the target pH was just met at the same dosing concentration of 20 g/L (JP). It is to be expected that the target can be achieved, despite the reduction of the contact time, if a higher concentration (40 g/L) is used. Technical rules in Germany for granular CaCO_3 filters specify a theoretical EBCT of 15–17 min for the given water quality at a temperature of 10 °C (DVGW-Arbeitsblatt W 214-2 (A) 2019). Even though the contact time found for the studied process is similar to that for a filter column, the required area for the proposed process combination of dosing and UF would still be significantly smaller. A specific advantage of the new process combination was that the recirculation of the backwash water provided additional time for the remaining calcium carbonate to react with the water and become dissolved.

A considerable difference in the filtrate quality between 20 and 40 g/L regardless of the retention time was not observed, demonstrating that the experiments and both dosing materials followed similar dissolution kinetics. This may be attributed to the fact that the kinetics significantly slow down as the system approaches equilibrium (Berner & Morse 1974; Plummer *et al.* 1978; Sjöberg & Rickard 1985), which seemed to be around pH 8.1–8.2 for the water tested. Furthermore, deacidification is controlled by the CO_2 concentration available rather than by the remaining amount of CaCO_3 , such as explained by the kinetic models of limestone filters (Yamauchi *et al.* 1987).

The change in water quality during deacidification processes may be visualized by plotting the CO_2 and HCO_3^- concentrations, as presented in Figure 2. The plot depicts the reference lines for different pH values, and on the upper left area, the initial concentrations of the respective experiments with JP as a dosing material. The change in these parameters toward the target pH closely followed the theoretical path for filtration over calcium carbonate (dotted line). As expected, the dissolution of CaCO_3 increased the bicarbonate and calcium concentrations from feed to filtrate (Table 2).

The calcium concentration could be increased by only 16–20 mg/L from the start to the end of the combined process, even though the available calcium concentration (as suspension) after dosing was around 160 mg/L for a dosing concentration of 20 g/L CaCO_3 (or 320 mg/L for 40 g/L). Nevertheless, this considerable overdosing of calcium carbonate is conditioned by the short-term filtration times and could be further minimized in long-term operations.

In contrast to the experiments with a short retention time (14 min with 30 L in the intermediate tank), the experiments with a longer retention time (34 min with 75 L in the intermediate tank) showed a slightly steeper slope than the theoretical path, as seen for 20 g/L and 75 L in Figure 2. This may also be a result of the abovementioned dependency of the deacidification process on the available CO_2 . Due to the longer retention time, more CO_2 may have been released into the air, and the HCO_3^- concentration, therefore, did not increase to the same extent as with shorter retention times. Likewise, the increase in conductivity and calcium concentration was slightly lower for the experiments with a 75 L intermediate tank volume.

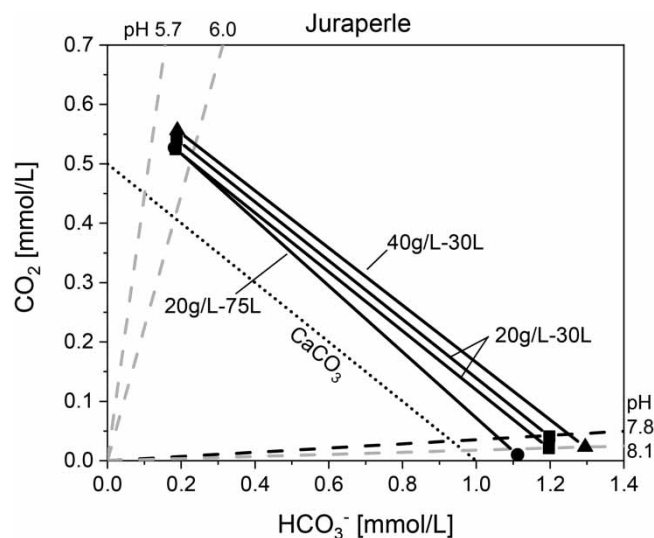


Figure 2 | Change in CO_2 and HCO_3^- concentrations for pilot plant experiments by a dosing of JP.

Fouling potential of CaCO₃

The TMP data collected during the pilot experiments provided limited information about the impact of the dosing material on the UF process. The plant was always operated at a single flux of 34 L m⁻² h⁻¹. The TMP showed no significant change throughout the whole experimental phase (total runtime of 40 h). This indicated to some extent that, at low fluxes and the concentrations tested, the UF operation may not be significantly impacted. Moreover, the membrane permeability may be sustained with the air-enhanced backwash measures implemented.

The first set of conditions evaluated for critical flux in the small lab-scale unit was that with a dosing concentration of 20 g/L KM and a volume of 75 L in the intermediate tank. The feed solutions were prepared to approximate a monitored turbidity of around 320 NTU. A series of stepping-flux experiments were conducted. However, no increase in the TMP was observed within the tested fluxes (50–220 L m⁻² h⁻¹). For this concentration, a critical flux was, therefore, not identified. Figure 3 shows the results of one of the experiments run at fluxes between 50 and 170 L m⁻² h⁻¹ at 60-min filtration-backwash cycles. From these runs with a dosing concentration of 20 g/L, a stable long-term operation may be expected.

The second set of conditions tested was that with a higher dosing concentration, namely 40 g/L JP and 30 L in the intermediate tank. For this case, the reference feed turbidity was expectedly higher with roughly 1,100 NTU. Already at the starting flux (50 L m⁻² h⁻¹), a considerable increase in TMP was observed. Therefore, filtration time was reduced to 15 min. As seen in Figure 4, the TMP increase in the example stepping-flux run for this concentration was more pronounced at the following flux increments. The repeated flux values showed a higher initial TMP as well as a slightly higher fouling rate (0.6 mbar/min larger for both 50 and 110 L m⁻² h⁻¹) compared with their respective filtration steps in the ascending phase, revealing the presence of an irreversible CaCO₃ layer on the membrane due to the higher CaCO₃ concentration in the UF feed.

The results obtained allowed for the derivation of the fouling rate at different fluxes, as presented in Figure 5. A critical flux around 55 L m⁻² h⁻¹ was determined for this dosing concentration. Beyond this clear threshold, fouling rates were significantly higher.

In order to gain an insight into how long a stable operation would be possible, an additional filtration experiment (Figure 6) was carried out over a longer operation time, such as in a conventional UF operation. The experiment was performed with a used membrane and under high CaCO₃-loading conditions (40 g/L JP). The flux was set to 70 L m⁻² h⁻¹ with the assumption that the operation is done within a flux range (50–90 L m⁻² h⁻¹), which is typical in practical applications (Lipp *et al.* 2005).

A linear increase in TMP was observed during all of the cycles, showing fouling formation throughout the entire run. A fouling rate of 1 mbar/min was estimated for the first cycle, followed by 0.7 mbar/min in the second. Afterward, the fouling rate was slightly more pronounced as the number of cycles increased, reaching 1.1 mbar/min for the last six cycles (after roughly 4.0–5.5 h runtime).

The backwashes carried out between filtration cycles were not able to recover the initial membrane properties, observably by the increasing initial TMP at every further cycle. The permeability gradually decreased after each cycle due to the remaining fouling layer. By the start of the last filtration cycle, around 20% of the permeability was lost.

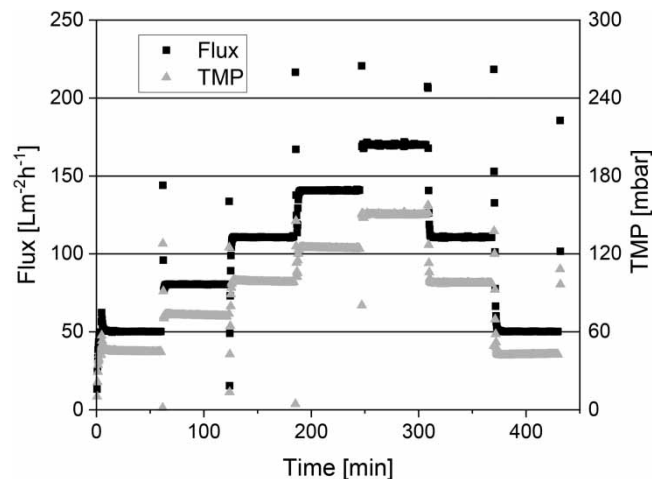


Figure 3 | TMP development during the stepping-flux method for 20 g/L KM at $\Delta t = 60$ min from 50 to 170 L m⁻² h⁻¹ by $\Delta J = 30$ L m⁻² h⁻¹.

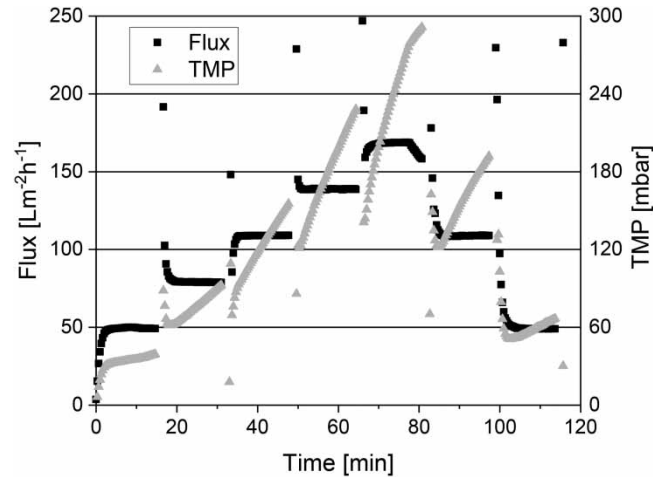


Figure 4 | TMP development during the stepping-flux method for 40 g/L JP at $\Delta t = 15$ min from 50 to 170 $\text{L m}^{-2} \text{h}^{-1}$ by $\Delta J = 30 \text{ L m}^{-2} \text{h}^{-1}$.

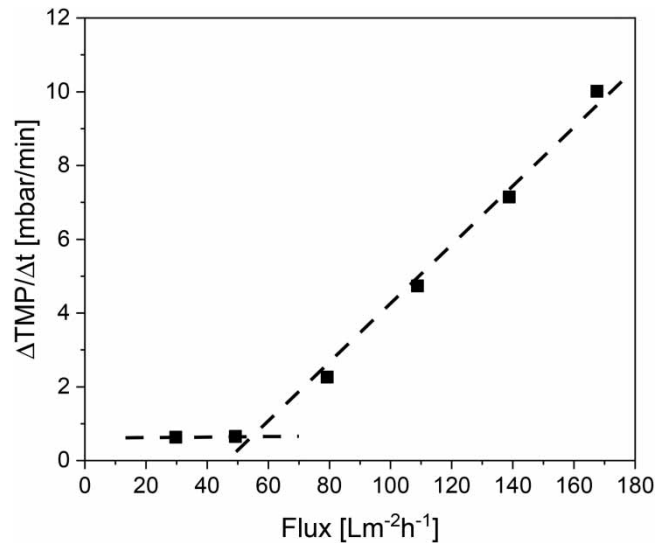


Figure 5 | Critical flux plot for 40 g/L JP with a 30 L intermediate tank.

A linear increase in TMP mainly predicts cake formation as a predominant blocking mechanism (Liu & Kim 2008; Iritani 2013). Although pore blocking mechanisms may also take place during dead-end filtration of particulate matter, especially in the early stage of operation (Wang & Tarabara 2008; Gao *et al.* 2011), this was not observed in the current experiment. One reason for this may be that the membrane was used in previous experiments and was only flushed and backwashed with demineralized water (not chemically cleaned). Another explanation may be that the CaCO_3 loading on the membrane was so high that the pore blocking phase was simply too short to be captured in the experiment.

Furthermore, the increasing fouling rate after every subsequent cycle suggested the possibility of a continuous compression of the CaCO_3 layer on the membrane surface (Boerlage *et al.* 2004). The presence of irreversible fouling may be problematic, assuming that the overall process combination is to be implemented in a real operation at these specific dosing conditions and at a flux greater than the determined critical value. While this operation behavior was to be expected with the knowledge of the stepping-flux runs, its investigation was relevant in order to determine the limits of the process.

Nevertheless, various maintenance measures may be put into practice to limit the impact, for instance, through the implementation of enhanced backwash procedures through air-scouring or at higher fluxes than the ones applied in the

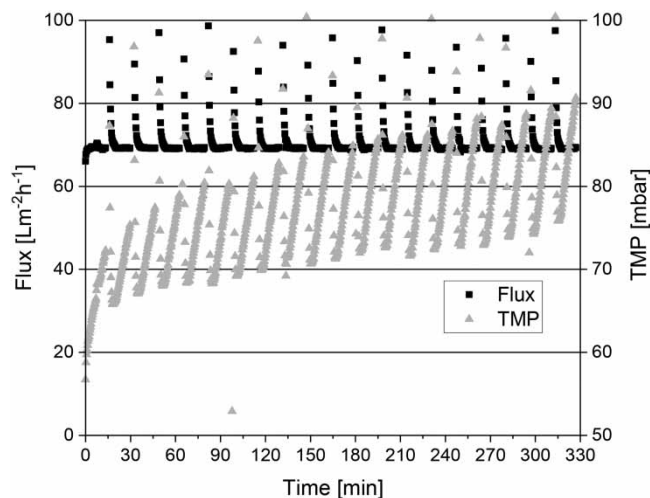


Figure 6 | Monitored TMP at a single-flux ($70 \text{ L m}^{-2} \text{ h}^{-1}$), 15-min filtration operation.

present small-scale filtration experiments. In addition, considering that the fouling layer consists of mineral deposits, regular acidic chemical cleaning may conceivably fully recover the initial permeability conditions (Lipp *et al.* 2016).

During the experiments with the pilot plant, only short filter runs were possible as the feed tank had a limited volume and synthetic water had to be prepared in batches. Another factor to consider is that the backwash water of the UF was directly returned to the feed. The future treatment concept is designed such that the backwash water of the UF process is collected in a storage tank and is treated by a second UF membrane and its filtrate will be returned into the feed tank. This concept could not be realized in the current study as the volume of backwash water was too small to operate a second UF membrane. Therefore, further research is planned with a larger pilot plant installed in the waterworks where spring water is available without volume restrictions.

CONCLUSIONS

The pilot plant experiments presented here showed that the combination of CaCO_3 -dosing and UF is a feasible process to treat soft water. Both the pilot- and the small-scale filtration experiments provided a broad understanding of the effect that the dosed material may have on the water characteristics and the UF performance through this process combination, which may be summarized in the following points:

1. When adding powdered calcium carbonate, the water quality changed in a similar manner as if the soft water was filtered over granular calcium carbonate, i.e., CO_2 concentration decreased, and pH and calcium concentration increased toward calcium carbonate equilibrium.
2. There was no difference in the reaction kinetics between the two tested materials with dosing concentrations of 20 and 40 g/L, respectively.
3. The volume of the intermediate tank could be decreased from 75 to 30 L (reducing the retention time from 35 to 14 min) and the concentration from 20 to 40 g/L simultaneously increased, while still achieving the target pH value and filtrate characteristics.
4. The pilot plant data collected while operating the UF module at a low flux rate of $34 \text{ L m}^{-2} \text{ h}^{-1}$ showed no noticeable impact in terms of TMP change at any of the dosing concentrations tested (15–40 g/L). The air-enhanced backwash process was, hence, sufficient to maintain the membrane permeability.
5. The short simulations of the dosing conditions revealed that CaCO_3 fouling was present only at the highest dosing concentration studied (40 g/L), at which operation over the determined critical flux suggested the formation of an irreversible layer. It may be implied that the extent of irreversible fouling depends on the filtration and backwash conditions.
6. The results showed that the desired filtrate quality was achieved under the prevailing conditions (i.e., temperature and raw water characteristics).

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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