

Review

Application of Magnesium Oxide Media for Remineralization and Removal of Divalent Metals in Drinking Water Treatment: A Review

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Abstract: The post-treatment of soft and desalinated waters is an integral step in the production of quality drinking water. Remineralization is therefore often essential in order to stabilize the effluent for distribution and to attain mineral levels that fulfill aesthetic and health goals. According to the World Health Organization, magnesium (Mg^{2+}) is a nutrient essential to human health. This review summarizes the effectiveness of magnesium oxide (MgO) media for soft water remineralization, as well as its potential for divalent metal removal (e.g., Mn, Cu, and Zn), which is of particular interest in small or residential applications. We present MgO sources, properties, and dissolution mechanisms. Water treatment applications are then reviewed, and the available design models are critically appraised in regard to remineralization and contaminant removal processes. In addition, we review the process operation challenges and costs. Finally, we discuss the use of MgO in combination with calcite and address the technical advantages and limitations compared to other available methods.

Keywords: magnesium oxide (MgO); post-treatment; water conditioning; mineral dissolution; contaminant removal; process modeling



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1. Background and Objectives

The quality of drinking water can vary from place to place, depending on the chemical, physical, biological, and radiological characteristics of the source water from which it is drawn and the treatment process [1]. In order to deliver safe and high-quality drinking water and minimize aesthetic concerns, the pH, alkalinity, and hardness of drinking water are among the parameters routinely monitored during the water treatment process; their interdependence also makes them of great importance when determining water treatment targets and operational constraints [2]. Accepted values of these parameters vary from one legislation to another. The World Health Organization references approximate guidelines for a drinking water pH between 6.5 and 9.5 and alkalinity and hardness of around 60–180 mg $CaCO_3/L$ [3].

Naturally soft surface waters and soft treated effluents from desalination processes have low pH, alkalinity, and hardness levels. These waters require an effective and cost-efficient method of post-treatment before distribution and consumption in order to achieve target parameter values. While these parameters are most often unregulated, soft water is corrosive and can degrade pipe surfaces, leaching metals and potentially causing health concerns regarding regulated contaminants such as copper and lead [3]. Additionally, corrosive water can have a metallic or acidic taste, as well as lead to complaints about staining [3]. The remineralization process, also called water conditioning or stabilization, typically aims to achieve bicarbonate equilibrium as well as to increase pH and alkalinity.

Previous studies have recommended a number of remineralization options at the municipal level [4]:

- The direct dosage of chemicals such as CO_2 , NaOH , $\text{Ca}(\text{OH})_2$, NaHCO_3 , Na_2CO_3 , CaCl_2 , MgCl_2 , and MgSO_4 ;
- Blending of hard water with a soft effluent;
- Mineral dissolution (e.g., calcite); or
- A combination of the abovementioned methods.

Amongst these options, the application of a passive mineral dissolution system at the point of entry (POE) is recommended for residential and small-scale water treatment systems [5]. Carbonate media, particularly calcite (i.e., CaCO_3) or limestone contactors, are common residential POE treatment methods due to a simple design that does not require continuous chemical addition while being inexpensive and easily available in the water treatment market [4]. Calcite contactors simultaneously increase carbonate alkalinity, calcium hardness, and pH [6]. Nonetheless, relying solely on calcite dissolution results in the addition of only Ca^{2+} to the treated water, whereas there is increasing human health-based evidence regarding the importance of Mg^{2+} in drinking water [7,8]. Inadequate total dietary intakes of calcium and magnesium are common worldwide. The World Health Organization has therefore suggested that the remineralization of drinking water supply take into account the nutritional requirements of the consumer, including calcium, magnesium, and other minerals based upon regional dietary composition [9]. Reduced cardiovascular mortality and other health benefits are associated with minimum levels of 20–30 mg/L calcium and 10 mg/L magnesium in drinking water [9]. Magnesium oxide (MgO , magnesia, or a commercially available product in the North American (NA) water treatment market, i.e., *CorosexTM*) can introduce Mg cations to the effluent and contributes to pH correction owing to the rapid dissolution and hydration of the oxide as $\text{Mg}(\text{OH})_2$ [10]. The main characteristics of the *CorosexTM* media are given in Table 1. Given the affordable price for a bag of 50 lb (< USD 100 in NA), a number of studies have also documented the successful application of MgO media for the removal of divalent metals from wastewater and contaminated groundwater [11–23]. Besides remineralization and the removal of divalent metals from aqueous solution, recently, Zhou and co-workers reported the efficient removal of natural organic matter from water via a hybrid nano- MgO c-microfiltration membrane process [24].

Table 1. *CorosexTM* media specifications reported in the literature [25].

Media	Purity (%)	Specific Gravity (g/cm^3)	Bulk Density (kg/cm^3)	Surface Area (m^2/g)
<i>CorosexTM</i>	98.0	3.60	1200	0.39

The technical and economic advantages of available remineralization and post-treatment options have previously been reviewed [4,26]. The aim of this review is to provide a comprehensive state-of-the-art summary of the effectiveness of MgO media in soft water remineralization for drinking purposes, as well as its potential for divalent metal removal (e.g., Mn, Cu, and Zn), an area that has, in our opinion, been overlooked to date. The combined mechanisms of remineralization and heavy metal removal are of particular interest for small-scale or residential applications, where the treatment installation should remain as simple and economically accessible as possible. Our paper is organized as follows. We present MgO sources and properties and dissolution mechanisms. Water treatment applications are then reviewed, and we critically appraise the models available for the design of remineralization and metal removal processes.

Magnesium Oxide Sources and Properties

MgO is a colorless to grayish-white inorganic compound that occurs rarely in nature as the pure mineral periclase [27]. When synthesized, the physical and chemical properties of

the mineral are primarily determined by the source of the precursor. MgO is often obtained from magnesite (MgCO_3), as well as from seawater or other brines that are rich in MgCl_2 , where lime (CaO) is added to the brine to produce a magnesium hydroxide (Mg(OH)_2) precipitate and CaCl_2 brine [28]. The magnesium oxide media is then dehydrated by a calcination process, from which a variety of MgO products with different properties and reactivities can be obtained [29]. The temperature and time of calcination are the main factors affecting the media particle surface area and reactivity.

Formed at lower temperatures, the media remains more porous and reactive (Figure 1) [29–35]. This lower temperature product is used in the water treatment industry due to its preferable reactivity and solubility [27].

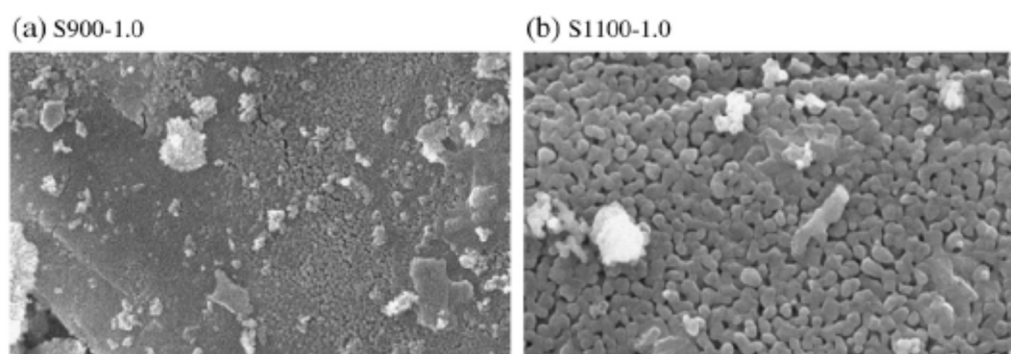
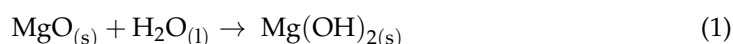


Figure 1. Effect of calcination temperature on the morphology of a MgO aggregate. The images show the result of 1 h of calcination at (a) 900 °C and (b) 1100 °C. According to the authors, specific surface area decreased by a factor of 13 for the sample calcined at only 200 °C higher [35].

2. Mechanisms in MgO Dissolution

2.1. Dissolution and Hydration Theory

For the purposes of reviewing the applications of MgO in the drinking water domain, this section discusses the main phenomena impacting MgO contactors, mainly: dissolution, hydration, precipitation, nucleation, and crystal growth. In water, MgO undergoes an alkaline reaction to form Mg(OH)_2 , also referred to as magnesium hydroxide, or brucite [36]:

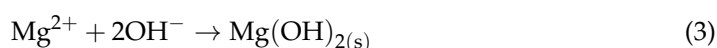


Two main dissolution mechanisms have been proposed in the literature [37–39]. The first is a shrinking core model applicable at temperatures ranging from 135 to 200 °C, which was modified to incorporate the influence of the hydration layer on the solid reactant [38,39]. For temperatures lower than 90 °C, which is the case for water treatment, a second mechanism is proposed. This mechanism is based on dissolution and precipitation steps, with some variation in the kinetic models and intermediate reactions. A compilation of the three main reaction pathways of the second model was summarized by Rocha and co-workers [40]:

- (1) Water adsorbs at the surface and diffuses inside porous MgO particles simultaneously;
- (2) Oxide dissolution occurs within particles, changing porosity with time:



- (3) Supersaturation, nucleation, and growth of magnesium hydroxide occur at the surface of the MgO particle:



Hydroxylation and dissolution of the media rapidly increase the concentration of Mg^{2+} and OH^- ions in solution, followed by a nucleation process or induction period [36,40,41].

Once the concentration of Mg^{2+} and OH^- ions in water has reached the saturation level, the precipitation (hydration) step begins [40]. Although Glasson suggested in 1963 that the continued dissolution of MgO is practically unimpeded by the hydration layer [42], others have claimed that the hydration process slows down further oxide dissolution and that the $\text{Mg}(\text{OH})_2$ layer becomes rate determining [36,37,39,40,43]. We therefore consider the latter dissolution–precipitation mechanism in this review, as it is more commonly observed in the literature.

Solubility has been cited to be as low as ~5 to 9 mg/L (~12 to 21 mg CaCO_3 eq/L) at 30 °C, but further conditions were not specified [18,27,44]. However, at room temperature and in acid-free desalinated water, solubility has also been measured to be as high as 34 mg/L (~84 mg CaCO_3 eq/L) [10]. Furthermore, although the oxide solubility is not found in reference texts such as *Aqueous Environmental Geochemistry*, the hydration product $\text{Mg}(\text{OH})_2$ is cited as having a significantly higher K_{sp} than that of calcite (Table 2) [45]. As it is generally accepted that MgO reacts to form $\text{Mg}(\text{OH})_2$ and that dissolution of the latter is rate determining, this second measurement appears to be more appropriate when compared to the known solubility of calcite (14 mg CaCO_3 /L).

Table 2. Solubility products at 25 °C and 1 bar total pressure.

Mineral	Formula	K_{sp} Expression	$-\log K_{\text{sp}}$
Brucite	$\text{Mg}(\text{OH})_2$	$[\text{Mg}^{2+}][\text{OH}^-]^2$	11.16
Calcite	CaCO_3	$[\text{Ca}^{2+}][\text{CO}_3^{2-}]$	8.48

In 2002, Birchall and co-workers proposed a dissolution–precipitation model for a range of temperatures and were able to validate it closer to room temperature (35 °C). Their kinetic model incorporated a factor of resistance as a result of the change in the porosity of media over time [37]. According to this model, $\text{Mg}(\text{OH})_2$ forms and deposits in the pores of the MgO particles, and the buildup of a hydroxide film increases the resistance of the media to further dissolution and hydration. This process is described by a semi-empirical equation that includes the mass balance of $\text{Mg}(\text{OH})_2$ produced during hydration [37].

$$C_{\text{Mg}(\text{OH})_2}(t) = C_{\text{MgO}}^0 - C_{\text{MgO}}^0 \exp\left[\frac{b}{a}(1 - e^{at})\right] \quad (4)$$

where $C_{\text{Mg}(\text{OH})_2}(t)$ is the concentration (mol/m^3) of the hydration product at a given time of reaction t (h), which is a function of the initial media concentration C_{MgO}^0 and its dissolution and hydration over time. The model also introduces the parameter a as a rate constant (h^{-1}) describing the variation in the porosity with time at a set temperature. Fitting transient experimental porosity data in the following equation obtains a :

$$\varepsilon(t) = \frac{\varepsilon^0}{\varepsilon^0 + (1 - \varepsilon^0) \exp(-at)} \quad (5)$$

where ε^0 and $\varepsilon(t)$ are the initial porosity and porosity at a given reaction time, respectively. The temperature dependence is included in parameter b :

$$b = \frac{\varepsilon^0 a_v}{1 - \varepsilon^0} k_0 \exp\left(-\frac{E}{RT}\right) \quad (6)$$

where a_v is the area per volume of the media (m^2/m^3), E is the reaction activation energy, R is the universal gas constant, T is the temperature (°C), and k_0 is the initial reaction constant ($\text{m}^4/(\text{mol}\cdot\text{h})$). Although the model was not able to predict the behavior of the authors' experimental samples hydrated below 35 °C, subsequent work confirmed that it well described experimental data for MgO in pure water at 30–90 °C [40,43].

The Johnson and Mehl, Avrami, and Kolmogorov (JMAK) model is a common approach used to describe nucleation and growth in the hydration process. This model assumes that nucleation is distributed randomly throughout the total volume of the reaction system [46–48]. However, hydration products have been found to form on the boundary of MgO grains [35,49]. A related boundary nucleation and growth (BNG) model incorporates such preferential nucleation [50]. Experimental results of MgO hydration have confirmed a better fit with an adapted form of the BNG over the JMAK model, especially at lower temperatures (20–50 °C) [51–53]. The BNG model can be written as:

$$C_{\text{Mg(OH)}_2}(t) = 1 - \exp \left[-2O_V^B \int_0^{Gt} (1 - \exp(-Y^e)) dy \right] \quad (7)$$

where $Y^e = \frac{\pi I_b}{3} G^2 t^3 \left[1 - \frac{3y^2}{G^2 t^2} + \frac{2y^3}{G^3 t^3} \right]$ (if $t > y/G$), and $Y^e = 0$ (if $t < y/G$)

G is the linear rate of growth of a product region in any direction, O_V^B is the boundary area per unit volume on which nucleation can occur, and I_b is the nucleation rate per unit area of the fresh MgO surface. The kinetics of the boundary nucleation process can be further described by two rate constants with units of inverse time [52]:

$$k_B = (I_b O_V^B)^{\frac{1}{4}} G^{\frac{3}{4}}; k_G = O_V^B G \quad (8)$$

The rate constant k_B describes the rate at which the particle surface is covered with the hydration product, whereas k_G describes the rate at which the hydration product fills pore spaces between the particles. If $k_B \gg k_G$, the boundary regions will be densely populated with nuclei, and the media will completely hydrate early in the overall process [52]. This scenario projects a thickening of the Mg(OH)_2 layer centered on the original MgO surface and predicts the hydration rate to decrease exponentially with time. However, if $k_B \ll k_G$ holds, the internal boundaries are expected to be only sparsely populated with nuclei. Media hydration would, therefore, occur essentially at the same rate in the entire system, as described by the standard Avrami equation [52].

From the literature, it is apparent that the models proposed by both Birchall and Thomas are able to adequately describe the dissolution and hydration of MgO powders. While the model presented by Birchall offers a numerically simpler representation of the hydration effects, only the modified BNG model has been verified at temperatures as low as 20 °C, which is the most representative of a water treatment context. Neither, however, has been applied to granular MgO. In a design context, it would therefore be crucial to verify that these theoretical models can simulate the dissolution and hydration behavior of a granular MgO column.

2.2. Parameters Affecting MgO Dissolution in Water Treatment

The overall rate of MgO dissolution and hydration is expected to be controlled by the diffusion of dissolved reactants and products or by surface reaction, depending on the system conditions [36,54–58]. The application of MgO in water treatment is therefore influenced by several factors, which we examine below.

2.2.1. Media Impurities

The rate of media dissolution is determined by the physical and chemical characteristics of the particle, including the type and the amount of impurities in the mineral composition [59]. In the presence of impurities, media dissolution may initially be controlled by more soluble fractions [12]. A number of researchers have suggested that CaO present in small quantities in caustic magnesia can initially control dissolution and the solution pH response until exhausted. In practice, we might expect this to result in a pH spike at the beginning of column operation before effluent pH stabilizes.

2.2.2. Particle Size and Shape

Particle size influences the media contact surface area and, consequently, the reaction surface between media and water. As particle size increases, the specific surface area of the particle in contact with water decreases, and extended contact times are required to attain the same degree of dissolution and hydration [60]. In a kinetic study of MgO dissolution, single-crystal surfaces exposed to aqueous environments for several days showed no evidence of hydration, while highly defective surfaces and powders with higher surface areas are readily converted to brucite [61]. The reactivity of MgO therefore appears to be directly linked to its size, shape, and specific surface area, which determine the adsorption sites available to interact with solution constituents [62]. Aside from dissolution and reactivity effects, particle size plays an important role when determining filter porosity and ensuring sufficient permeability of media in an engineering application [15]. Commercially, the effective particle size (D_{10}) of granular MgO sold for NSF-approved drinking water treatment ranges from approximately 1.2 to 3.5 mm.

2.2.3. Internal Particle Porosity

MgO media consists of a solid matrix with interconnected voids, henceforth referred to as particle porosity. Particle porosity can be generally described as the ratio of void-space volume (V_v) to bulk particle volume (V_T) [63]:

$$\varepsilon = \frac{V_v}{V_T} \quad (9)$$

This should not be interpreted as the porosity of the filter bed, which considers the volume of voids between particles and contributes to the permeability of the filter [64]. Particle porosity is known to be positively correlated to specific surface area (SSA), which is directly proportional to the hydration degree of MgO particles [39,65]. As a result, MgO reactivity decreases remarkably with lower SSA [35,65]. In 2014, Jin and Al-Tabbaa observed a hydration limit due to the incomplete hydration of the most interior area of magnesia particles [65]. They found that the hydration degree increases linearly with the increase in SSA before this limit is reached [65]. Alteration of initial particle porosity caused by the precipitation of $Mg(OH)_2$ has also been reported in the literature [40,43]. As the reaction progresses, both the surface and pores of magnesia particles are progressively covered by a hydroxide film, changing the porosity of the media [40]. Accordingly, the diffusion of water is hindered inside particles, reducing the overall reaction rate through diffusive control [40]. In practice, this phenomenon implies that pH and alkalinity are not expected to be stable over the cycle of operation of a contactor, with higher values predicted with fresh media, while long-term dissolution is controlled by the dissolution of $Mg(OH)_2$.

2.2.4. Feed Water Composition

Acidity (pH)

Kinetic theories concerning the dissolution of ionic oxides predict that their dissolution rate exhibits an inverse exponential dependence on solution pH [36,57,66,67]. In 1969, Vermilyea proposed a similar method of controlling MgO dissolution by proton attack at low pH and proton diffusion at higher pH [36]. Fruhwirth and co-workers studied rate-controlling processes by conductivity and scanning electron microscopy measurements during and after hydration experiments. They found that the hydration rate of MgO is controlled by the dissolution rate, with the overall reaction being limited by H^+ and OH^- diffusion [54].

MgO Saturation Index

The MgO saturation state of the solution in contact with MgO media and hydrated $Mg(OH)_2$ surfaces largely controls the extent of media dissolution. In general, the saturation index (SI) of a mineral phase can be calculated using the ion activity product (IAP) and solubility constant K [63]:

$$SI = \log(IAP/K) \quad (10)$$

Saturation is influenced by pH and temperature and, more specifically, the difference between the solution pH and the saturation pH (pH_{eq}), which can be calculated for a system at a given temperature [68]. When $SI < 0$ or the solution pH is less than the calculated equilibrium pH, the mineral phase is expected to dissolve, as the solution is undersaturated. Precipitation occurs when $SI > 0$, or when the solution pH exceeds pH_{eq} , is supersaturated with respect to the mineral phase [63]. Furthermore, SI dictates if precipitation will be homogeneous (high SI) or heterogeneous (low SI). These equilibrium principles will govern the extent of the dissolution of MgO in water treatment, with influent pH and temperature acting as important factors in determining the remineralization capacity of the media.

Temperature

The temperature dependence of the MgO hydration reaction rate constant is consistent with an Arrhenius-type formulation [54,69,70]. The solubility of the hydration product $Mg(OH)_2$ has been found to decrease as the temperature of the system increases [36,40]. Consequently, MgO media experiences accelerated hydration and reduced solubility with increasing temperature [42,43,53,71,72]. It should be noted that high supersaturation and nucleation rates were found to be reached rapidly within a few minutes, even at a temperature of 35 °C [40]. This is of particular interest because it suggests that at low temperature, $Mg(OH)_2$ precipitates as small particles, which form a very porous agglomerate [40]. Moreover, the morphology and distribution of the $Mg(OH)_2$ hydration product appear to change with temperature, which in turn impacts the reaction kinetics [53]. However, the effect of temperature on the dissolution and hydration of granular MgO at room temperature currently remains overlooked and warrants further investigation.

Aqueous CO₂ Content

The maximum magnesium ion concentration that can be achieved by magnesia dissolution in acid-free soft water is limited by its low equilibrium solubility of 34 mg/L (84 mg $CaCO_3$ /L) [4]. The extent and rate of MgO dissolution can be enhanced by acidifying the inlet solution with CO₂. In water, MgO and $Mg(OH)_2$ react at the same rate with aqueous CO₂ [73]. These similar rates could be explained by the fact that media dissolution is inhibited by the $Mg(OH)_2$ hydration layer present on the surface of MgO, which further confirms the rapid formation of a rate-controlling hydration film [39]. In addition to shifting the system solubility equilibrium, the presence of available dissolved CO₂ can promote the precipitation of magnesite ($MgCO_3$) or partially recrystallize the hydration layer to hydromagnesite rather than solely hydrating MgO as $Mg(OH)_2$ in pure water [74]. Figure 2 compares the theoretical solubility of calcite or magnesium oxide in pure water at 25 °C. We assume that the hydration of MgO occurs such that dissolution is rate-limited by the hydration product $Mg(OH)_2$ and use the K_{sp} values given in Table 1.

Solid lines are simulated in a closed system, where the influent is not equilibrated with CO₂ (g) before entering the batch simulation. The respective $CaCO_3$ and MgO hardness release can be seen to increase significantly with influent alkalinity in Figure 2A, which indicates the importance of dissolved CO₂ in media solubility. The simulated variation in media solubility and consequent hardness response for influents exposed only to atmospheric CO₂ (410 ppm) appears to be minimal (2 mg $CaCO_3$ /L) in comparison. It should be noted that alkalinity varies significantly across the pH range described by this simulation. For example, the addition of 25 $CaCO_3$ /L alkalinity as 0.0006 M $NaHCO_3$ results in alkalinities ranging from 10 to 30 along the same closed system curve. Figure 2B further suggests that the pH correction and remineralization capacity of $CaCO_3$ is more sensitive to the presence of dissolved CO₂ than that of MgO. In the closed-system simulations, we see that increasing influent alkalinity increases the effluent pH range as well as the range of effluent hardness levels. While MgO also experiences improved hardness release in response to increasing concentrations of CO_{2(aq)}, its pH response remains more stable. In pure water and a closed system, the simulation data appear as a cluster of points rather than a curve. This confirms that without the presence of CO_{2(aq)}, both media experience a very narrow dissolution

range and consequently produce an effluent with more controlled pH correction. In a treatment context, $\text{CO}_2(\text{aq})$ therefore remains of great importance not only in optimizing the remineralization capacity of the media but also in understanding the sensitivity of effluent water quality to variation in influent $\text{CO}_2(\text{aq})$.

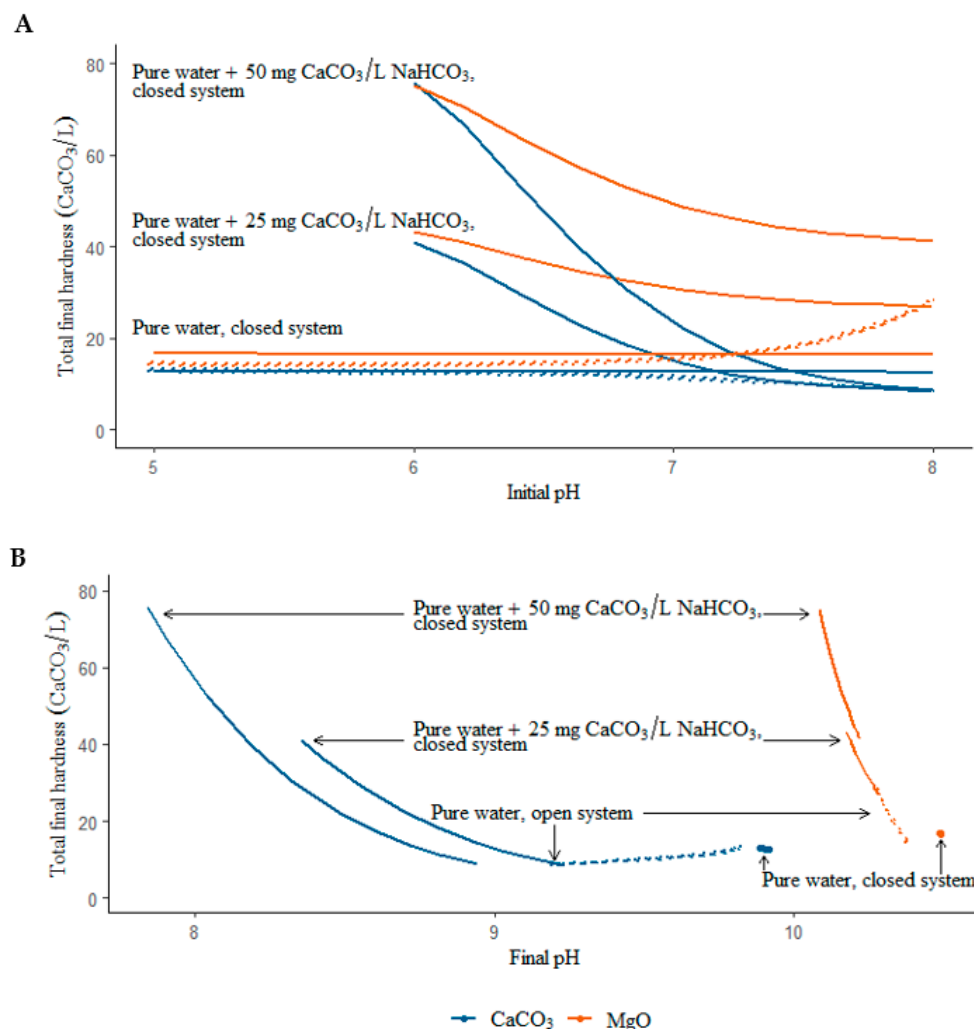


Figure 2. (A) Theoretical solubility curves for pure CaCO_3 or MgO in open (dotted) and closed (solid) systems. Before entering the batch simulation, the influent has zero Ca^{2+} or Mg^{2+} hardness and is prepared at varying pH by the addition of HCl or NaOH . (B) Corresponding effluent pH response to the dissolution of each media.

Presence of Anions

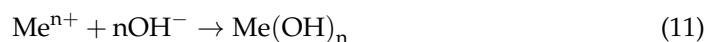
The presence of anions other than OH^- can form a hydration barrier by competing with OH^- for the positively charged MgOH^+ surface during the dissolution process (Amaral et al., 2010). The authors reported a delay in the MgO hydration reaction when CaCl_2 was added to the solution [71]. However, if the anion competition results from the dissolution of a magnesium salt (i.e., MgCl_2), the common-ion effect increases the Mg^{2+} concentration. Rather than delaying hydration due to a protective anion layer, magnesium supersaturation is accelerated and the precipitation of brucite is more effective [71]. Vermilyea also noted that certain dissolution accelerators such as phosphate can form protonated ions that react directly with the surface hydroxyl and, subsequently, increase brucite dissolution [36]. However, in the context of soft water treatment, we do not expect the concentration of anions to be significant enough to impact the dissolution of MgO media.

Presence of Dissolved Metals

Dissolved metals can form local, unreactive MeOOH networks on oxide and carbonate surfaces [68]. Among divalent metals (Me^{2+}), Pb and Ca cations promoted $\text{Mg}(\text{OH})_2$ dissolution, and Co and Ni inhibited dissolution rates at room temperature and slightly acidic pH (4.9) [75]. In a water treatment context, the interaction of dissolved metals with a mineral surface becomes interesting if it leads to the retention of the metal. Using calcite for the remineralization and simultaneous removal of manganese from synthetic feedwater, Pourahmad and co-workers found that the media was progressively loaded with a stable MnO_x film [76]. The authors proposed that, upon initial removal as MnCO_3 through an ion-exchange sorption reaction, the metal carbonate slowly recrystallizes as MnO_2 . The authors also observed improved metal removal due to the autocatalytic nature of the adsorption and oxidation of remaining dissolved manganese by the oxide film. MgO is also known as an effective media in metal removal and retention, as discussed in further detail below.

Precipitation

The most common method for removing metals from solution is by adjusting the pH in order to precipitate the metal as its hydroxide (Shand, 2006b):



Although the dissolution of carbonates allows for the removal of trivalent metals through precipitation, efficient divalent metal removal (i.e., Zn, Mn, Cu, Pb, Ni, Co, and Cd) requires a higher pH range (9–10) produced by the dissolution of MgO [12]. The solubility and surface charge of MgO also favor the formation of compact precipitates, as the precipitation reaction occurs at the positively charged hydroxylated surface, whereas the formation of a hydroxide suspension would mainly take place in more soluble bases [18].

Sorption

The difference between MgO metal removal by sorption and precipitation is not thoroughly addressed in the literature. Studies on $\text{Me}(\text{OH})_x\text{-H}_2\text{O}$ interactions suggest that hydroxide particles can remove metals within a periphery of localized high pH close to the particle surface, even in cases where the bulk solution pH does not precipitate metals from solution [19,77]. Other researchers have suggested that the dissolution of MgO promotes the oxidation of metal hydroxides into negatively charged metal oxides, which sorb metal cations remaining in solution [15,17,78–80]. As a combined mechanism, metal removal could therefore be considered a sorption–coprecipitation process [15].

Cation Exchange

Cao and co-workers suggested a direct cation exchange between the magnesium cation and the divalent contaminant ion rather than removal by sorption to the oxidized metal hydroxide. They justified this mechanism by the quantitative measurement of the molar amount of Mg^{2+} released and the amount of Pb^{2+} or Cd^{2+} adsorbed in batch experiments, finding a linear relationship [79]. While we were not able to find any other work investigating metal removal by cation exchange for MgO, previous research has found that calcite effectively removes Me^{2+} from solution by such a mechanism [76].

2.3. Comparison to Calcite Dissolution

There are several theoretical models for calcite dissolution, most importantly including those of Plummer, Parkhurst, and Wigley (PWP); Yamauchi; and Letterman [5,81,82]. In the Yamauchi and PWP models, the phenomenon of diffusional mass transport is neglected in favor of surface reactions, while Letterman assigns mass transfer as the key controlling mechanism in predicting calcite dissolution. Modifications have been proposed by several researchers to better simulate contactor experimental data [83–86].

Although reviewing the CaCO_3 system would be beyond the scope of this work, more in-depth reviews can be found elsewhere: [87–89]. As CaCO_3 is a prevalent media used within the treatment context reviewed here, it is worth mentioning that the parameters discussed in Section 2.2 exhibit the same tendency to stimulate or hinder the dissolution of both MgO and CaCO_3 minerals. This is important for engineering applications, as it allows for the design to optimize the dissolution of both media without hindering the other. However, while changes in these parameters produce similar impacts on dissolution for MgO and CaCO_3 , the specifics of the media response remain different. Notably, both media experience increased dissolution rates at lower pH (Figure 2), whereas their mineral phases stabilize at different equilibrium pH [45]. This is especially important to consider in the water treatment context, as pH can significantly impact metal removal. In addition, a mixed CaCO_3 – MgO contactor will be unable to attain equilibrium concentrations for both minerals, which will impact respective Ca^{2+} and Mg^{2+} remineralization [90]. Both aspects are discussed in detail in the following section on water treatment applications.

3. Water Treatment Applications

In drinking water systems, remineralization is often implemented as a polishing step at the end of the treatment chain. This is the case for naturally soft ground- or surface waters and desalinated waters. For treated water, the effluent may require remineralization following non-selective processes such as reverse osmosis (RO) and other membrane applications such as nano-filtration, which deplete hardness and alkalinity. Importantly, most conventional post-treatment technologies do not enrich water with Mg^{2+} , especially at the smaller, domestic scale [4]. Mineral sources of Mg^{2+} in the water treatment industry include dolomite ($\text{CaMg}(\text{CO}_3)_2$) and MgO media. Although not explicitly recommended in the literature, it appears that it may be advantageous to implement a CaCO_3 – MgO blend rather than using $\text{CaMg}(\text{CO}_3)_2$. For example, Luptáková and co-workers found that magnesium content dominated in treated water when using half-calcined dolomite and suggested the use of dolomite with higher Ca content for more balanced effluent target values [91]. However, dolomite is known to be naturally more heterogeneous than limestone [92]. This requires a dolomite-based remineralization system to exhibit greater design flexibility compared to a conventional calcite contactor [4]. Most literature reports based on laboratory batch tests also indicate that dolomite dissolution is much slower compared to that of calcite, while MgO is known to be significantly more soluble than CaCO_3 [84,93,94]. In addition to preferable reaction kinetics, the use of MgO mixed with CaCO_3 rather than solely dolomitic media could offer greater adaptability of the Ca–Mg remineralization ratio as well as improved control of pH adjustment needed for divalent metal removal.

In drinking water treatment, it is recommended to use chemicals that are NSF International (NSF) or American National Standards Institute (ANSI) certified. *NSF/ANSI Standard—60 for Drinking Water Treatment Chemicals* applies to process media accepted in water treatment and distribution systems. In North America, Martin Marietta Magnesia is the only *NSF/ANSI Standard 60* supplier of MgO media products for pH adjustment or precipitation processes.

3.1. Main Applications

3.1.1. pH and Alkalinity Correction

Waters with low pH and alkalinity are considered soft and corrosive, while a high pH level is often associated with a higher mineral content in the form of alkalinity and hardness. MgO dissolves readily in aqueous environments, and it is the hydration product $\text{Mg}(\text{OH})_2$ that is considered to control the residual effluent pH and alkalinity. The $\text{Mg}(\text{OH})_2$ film releases OH^- slowly over time due to its low aqueous solubility [17,95]. Following an initial rapid pH increase with MgO dissolution, the stabilized effluent pH is therefore constrained by $\text{Mg}(\text{OH})_2$ dissolution, providing a long-term source of OH^- alkalinity at a pH range of 9–11 [11,15,96,97]. Compared to operating a conventional limestone contactor,

authors have found that in column experiments of comparable dimensions, MgO media was able to remove twice the acidity (in CaCO_3 equivalents) [17,98]. Figure 3 summarizes the main reaction mechanisms that occur in the treatment applications of MgO and calcite. For drinking water applications, the application of MgO on its own risks producing ‘over-corrected’ water. The media is therefore of more interest as an addition to the standard calcite contactor, moderating the pH correction as well as producing remineralized water with both Ca^{2+} and Mg^{2+} hardness.

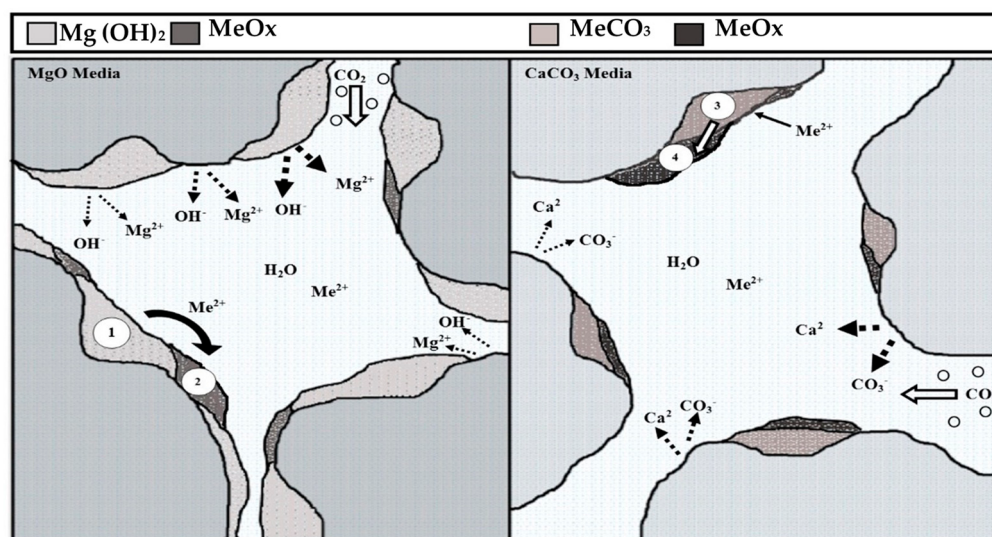


Figure 3. Comparative summary of MgO and CaCO_3 remineralisation in soft water containing dissolved divalent metals. The thickness of the dashed arrows indicates relative reaction kinetics, and the presence of CO_2 increases the dissolution of the two media and the MgO hydration product, $\text{Mg}(\text{OH})_2$. The main reactions in each system are as follows: (1) rapid hydration of MgO into $\text{Mg}(\text{OH})_2$, and (2) uptake of Me^{2+} by a sorption-coprecipitation process or possible cation exchange. In a CaCO_3 system the Me^{2+} is removed by (3) ionic exchange, followed by (4) the slow recrystallisation of MeCO_3 to MeO_2 .

3.1.2. Magnesium Hardness

In the last decade, guideline recommendations have been adapted to include the importance of Mg^{2+} alongside Ca^{2+} in the potential health benefits of mineralized drinking water [8]. Since then, several workers have adapted treatment chains to include magnesium addition to the water conditioning process. However, this includes only a few who aimed to introduce magnesium hardness through the application of MgO media [10,76,99]. Both Hasson and Schwartz focused on Mg^{2+} remineralization, the latter improving the dissolution process with either CO_2 or H_2SO_4 injections at the inlet of MgO columns [10,99]. Working at a scale more appropriate for domestic application, Pourahmad and co-workers successfully adapted a pure calcite contactor to improve the long-term remineralization of drinking water by introducing a fraction of MgO into the column [76]. It appears that despite being a media well known to the industry, the investigation of MgO remineralization for drinking water purposes has been somewhat overlooked in the research field.

3.1.3. Metal Removal

While there is relatively little literature addressing the use of MgO for metal removal in drinking water, it is well established in the field of wastewater and contaminated groundwater treatment. MgO powder is used as an alkaline material to facilitate oxidative precipitation, followed by coagulation and flocculation in wastewater treatment applications [100]. Cortina and co-workers showed that MgO satisfies the conditions for application as a passive in situ reactive barrier (PRB) for groundwater treatment, notably due to satisfactory reactivity, permeability, and durability [12]. Conventional active treatments typically rely

on the continual addition of energy and/or chemical reagents to remove metals, which is costly [101]. While passive treatments are less precise and require careful design and characterization of the influent to be treated, they generally require lower capital, maintenance, and operating costs than an active system [102]. The same arguments that support the use of MgO as a geochemical passive treatment option in the wastewater industry make it appealing for use in smaller systems in the drinking water industry. In these systems, the media reduces metal concentrations by precipitation and sorption mechanisms while requiring minimal maintenance.

Efficient removal of divalent metals has been incorporated into multi- and single-step passive treatment systems using reactive tanks, columns, and permeable reactive barriers [11,13–19,98,103,104]. In a large number of these studies, the long-term operation of a MgO-based metal removal technology eventually suffered from reduced performance due to media coating and decreased permeability. It was noted that coarser-grained (2–4 mm) MgO columns were much less reactive and exhibited a reduced rate of pH adjustment and dissolved metal removal [60].

3.2. Process Operation

3.2.1. Scale Formation

In the water treatment industry, scaling can become an issue both during the treatment operation and during the distribution of the treated effluent. Scaling occurs when the pH, temperature, and mineral content conditions are such that saturation thresholds are exceeded and minerals precipitate, as discussed in Section 2. The deposition of $\text{Mg}(\text{OH})_2$ can be promoted by an increase in temperature or pH or a decline in dissolved CO_2 [105]. It is therefore critical to monitor these parameters in order to avoid unwanted scaling. Distribution networks that include water heaters are of specific interest, as treated water is subjected to a significant rise in temperature. For magnesium, historical data obtained from studies where magnesium hydroxide precipitation in hot-water tanks and lines was problematic suggest that an effluent at 24 °C should have a pH of less than 9.0 and a magnesium hardness of less than 40 mg CaCO_3/L in order to avoid scaling in water heaters [106]. In practice, the calculation of saturation indices is used to predict the tendency and extent of precipitation. Furthermore, treated water is unlikely to contain only magnesium hardness. Predictive tools such as that proposed by Bahadori (2010) offer an approach to assess operational issues and include the magnesium contribution to estimate scaling [107].

3.2.2. Issues with Filtration Media Coating

As mentioned previously, several authors have noted a performance decrease in MgO water treatment systems due to reduced system permeability as a result of media coating. While initial studies performed by Navarro and co-workers did not find targeted metal removal to be affected by the presence of other metals, other workers later found that aluminum (Al) and iron (Fe) should preferably be removed prior to MgO water treatment in order to prevent prematurely clogging the column [11,15,17]. These authors also noted a decrease in column efficiency due to media coating and passivation when only a small portion of the reagent had been consumed [17,98]. The results also showed that grain size selection is an important factor influencing the extent of dissolution, and that the proportion of MgO when mixing with another media needs to be adapted to extend operation lengths. Notably, the above column experiments investigated the treatment of acid mine drainage (AMD) with high inlet contaminant concentrations of up to 300 mg/L aluminum and 100 mg/L iron [17,98]. The contaminant concentrations in these studies significantly exceeded the concentrations expected in drinking water sources.

The operational lifespan of a treatment relying on MgO dissolution has been improved by mixing the alkaline material with a larger inert matrix, such as quartz sand or wood chips. This allows for the reduction in MgO grain size, increasing reactivity and specific surface in order to retard passivation while the system permeability remains high [12,60]. The use of fine-grained media also allows for more complete dissolution before the growing

layer of precipitates passivates the particle. Over a year, it was found that despite a substantial decrease in hydraulic conductivity, finer-grained column experiments treating high zinc and manganese concentrations did not clog at any time [60]. In drinking water treatment, interference from high metal concentrations would not be expected once at the point of remineralization. However, it remains interesting to note that within the range of maximum acceptable concentrations allowed for Al (2.9 mg/L) and Mn (0.12 mg/L) [108], a MgO column may be able to further reduce regulated contaminant levels.

3.2.3. Past Attempts to Model Process Performance

The process design of a MgO-based drinking water treatment depends on the scale of operation and requires (as most treatments do) a design based on specific treatment targets and influent water characteristics. Modeling can be useful to verify design robustness and to predict the operational capacity and lifespan of a given scenario.

To the best of our knowledge, there is currently no commercial software available to guide the design of a MgO or blended MgO/calcite contactor for drinking water applications. The models presented in this section therefore concern other types of water treatment applications and are restricted to fixed-bed, up-flow granular column experiments, as such a configuration is the most common option for MgO application in drinking water treatment. Furthermore, all models consider the dissolution of $\text{Mg}(\text{OH})_2$ to be rate controlling, since the MgO media is considered to hydrate almost instantaneously. There are four main phenomena of interest when simulating column treatment:

- (1) Dissolution;
- (2) Particle evolution and passivation;
- (3) Contaminant removal; and
- (4) Permeability loss.

Table 3 summarizes the main approaches to modeling MgO column treatment, according to the phenomena listed above. While several workers have successfully modeled this system, their focus varies depending on the treatment objective and never encompasses all four phenomena. For example, a reactive transport model was able to simulate measured depth profiles and was used to extrapolate the lifespan of a MgO column applied for divalent metal removal [14,60]. Other authors were also able to reproduce laboratory results for metal retention through the geochemical modeling of saturation indices and comparison with confirmed mineral phases along the depth profile of MgO columns [11,12,15,98]. These latter works do not, however, provide complete design models for the engineering application of MgO treatments. Courcelles and co-workers estimated the longevity of PRBs using MgO column laboratory experiments, coupling chemical reactions and principles of transport in porous media, and incorporating system pore volume evolution due to precipitation [13]. Due to the treatment context, their work focused on the impact of permeability reduction on hydraulic conductivity rather than on maintaining target effluent concentrations. The authors pointed out that there are several model formulations that consider column porosity loss but neglect the evolution of SSA. In doing so, these approaches underestimate the loss of permeability caused by changes in pore geometry. It was therefore proposed that modeling SSA is essential to understanding the precipitation-clogging phenomenon, and that this factor can be included using the Kozeny–Carman equation in conjunction with the floating spheres model [13].

Table 3. Models of MgO in water treatment applications.

Authors	Modeled Phenomenon	Validation
Dissolution in water		
59, 14	$r = 10^{-4} \cdot a_{H^+}^{0.45} \text{mol} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$	A, B
10, 99	$m = A r (1 - \Omega)$ $\frac{C_2}{C_s} = 1 - \exp\left(-\frac{K \cdot a \cdot Z}{L}\right)$ $r = \frac{Q \cdot d[Mg_T]}{dS_p} = k\{[Mg_T]_e - [Mg]\}$ $\ln \frac{[Mg_T]_e - [Mg_T]_L}{[Mg_T]_e - [Mg_T]_0} = \frac{6k}{\varnothing} \cdot \frac{(1-\varepsilon)}{d_p} \cdot \tau$	B
Particle evolution and passivation		
59	$A = A_0(V/V_0)^{2/3}$	None
Contaminant removal		
59, 14	$-\frac{d[Mn^{2+}]}{dt} = k'_1 [O_{2(aq)}] [OH^-]^2 [Mn^{2+}] + k'_2 [O_{2(aq)}] [OH^-]^2 [MnOx] [Mn^{2+}]$	A, B
13	$\frac{\partial q}{\partial t} = A \cdot e^{-\left(\frac{E_a}{RT}\right)} = \Pi_i a_i^{\eta_i} (1 - \Omega^p)^q \cdot Sv$	B
Permeability and hydraulic conductivity loss		
13	$k = \frac{\varnothing^3}{\tau \cdot (1-\varnothing)^2 \left(\sum_{i=1}^{N_m} \frac{\varphi_i}{r_i}\right)^2}$ $\varnothing = 1 - \sum_{i=1}^{N_m} \varphi_i$	C

A Verified by mineralogical analysis (XRD) or (EPMA); B Verified by tracking the evolution of dissolved constituent concentrations; C Verified by recording differential pressure measured along the depth profile.

Although the aims of the aforementioned laboratory- and pilot-scale studies were to implement a MgO passive low-flow treatment for AMD or contaminated groundwater, the authors monitored and successfully simulated parameters that are equally important to the drinking water area, such as pH, divalent metal retention, the evolution of magnesium and calcium concentrations, and changes in column permeability.

Hasson, Schwartz, and co-workers investigated the remineralization of desalinated waters with MgO pellets in pure water or waters acidified with CO₂ and H₂SO₄ [10,99]. While both of their models were able to simulate measured Mg²⁺ addition, they did not address the operational lifespan of the column beyond the immediate remineralization demands. Both models consider the kinetics of MgO dissolution to be mass transfer controlled. The modification of the dissolution expression in acidified solutions is not included in Table 2 but can be found in greater detail in Schwartz et al. [99].

Notably, only one model [13] incorporated the change in the reactivity of a column due to the evolution of particle porosity and specific surface area. However, based on previous work conducted investigating the hydration of MgO, the transient behavior of MgO particle porosity could have a significant effect on the hydration and the passivation of media.

To summarize, the four main phenomena listed above are collectively addressed by the design models in Table 2; however, none specifically target both the simultaneous remineralization and metal removal capacity of the media, which is of interest for drinking water applications. Their validation is therefore currently limited to either permeability loss, remineralization, or metal removal breakthrough when modeling long-term operation. While not yet adapted to MgO media, Pourahmad and co-workers investigated CaCO₃/MgO blends in drinking water treatment and successfully validated manganese removal and remineralization via PHREEQC for a pure calcite column [76]. Importantly, this work suggests that although media coating eventually passivates the grains in terms of hardness and alkalinity release, this same coating improves metal removal due to the autocatalytic sorption of manganese by MnO_x [76]. This illustrates the need to further investigate the modeling of media evolution over time when aiming to maintain both hardness addition and metal removal objectives.

3.2.4. Costs

Costs associated with post-treatment are site-specific and vary depending on the target effluent quality, as well as the technology chosen and the additional costs of consumables. The costs of implementing calcium-based remineralization processes such as lime/ CO_2 and CaCO_3 dissolution at a municipal scale have been previously discussed [4,109]. While data are widely available concerning the application of chemical dosing and CaCO_3 -based remineralization, they are less so for a MgO or mixed CaCO_3 /MgO remineralization step. At the residential scale, the chemical dosage of Mg^{2+} using MgCl_2 or MgSO_4 is economically and technically uninteresting, as the aim is to design an affordable and simple treatment chain, minimizing human intervention. In this context, the application of MgO media for simultaneous remineralization and metal removal is of particular interest, as it has the potential to simplify the treatment design. However, the lack of data regarding metal removal by MgO in drinking water applications is currently a limiting factor in predicting operational costs. Furthermore, the cost-effectiveness of implementing a mineral contactor is directly linked to its operational longevity. This aspect emphasizes the importance of developing a design model for MgO remineralization and metal removal. As a baseline, it is worth noting that NSF/ANSI Standard 60 certified MgO media currently (2021) retails at a slightly higher cost (5.73 USD/kg) than calcite (4.44 USD/kg) and dolomite (5.38 USD/kg) per 50 lbs. However, commercial sellers recommend the addition of only 10–20% MgO content, which translates to a relatively small increase in material costs when introducing MgO to the treatment design.

4. Discussion and Suggestions for Further Research

It is worth mentioning that this review relies on some work outside the drinking water literature due to the surprisingly limited research concerning MgO in drinking water applications, despite being a common media used in the industry. Accordingly, it will be important to confirm that the phenomenon and performance described by authors working with scenarios of high contaminant concentrations are applicable to the lower concentrations found in drinking water systems. Furthermore, several of the above studies were performed at laboratory scales in controlled environments, and a degree of discrepancy between theory and practice must therefore be expected.

Validated design models assist in the application of treatments in the real-world context. In this sense, it becomes especially important that MgO dissolution theory is effectively translated to engineering applications. The following section discusses aspects that we believe could be strengthened by further investigation, particularly concerning MgO application for the removal of regulated contaminants.

The models discussed in Section 2.1 are derived from MgO dissolution theory outside of the water treatment realm. It should be noted that in what little literature could be found on MgO in drinking water treatment, the kinetics of the media dissolution is most often treated as a simple mass transfer process. While measurements of granular MgO dissolution are shown to be in excellent agreement with values predicted from such a kinetic model [4,10], it cannot adequately predict the longevity of the system if we suppose that the transient behavior of the porosity of MgO and the passivation of its surface are two important factors influencing dissolution kinetics over time. Although it has been noted by several authors and was incorporated into the design model presented by Courcelles et al. (2011) for an in situ GW treatment application, this aspect requires more attention in drinking water applications [13]. The evolution of MgO media reactivity is integral to the operational lifespan of the treatment, not only regarding cost but also in order to guarantee effective treatment. If installed domestically for metal removal without being subject to the same rigor of process control available at the municipal level, early media passivation could lead to the unintended consumption of drinking water that has close-to-pre-treatment contaminant levels. Furthermore, the longevity of metal retention in a MgO system remains unclear. We know, for example, that the leaching of metals is pH-dependent, and while the metal hydroxides precipitated in the treatment

process should remain stable within the pH range 9–11, a decline in media reactivity over time could result in the release of previously removed contaminants as the system pH decreases. Therefore, it is essential that we determine the dominant process responsible for the removal and retention of divalent metals when working with MgO in the context of soft water remineralization. Considering that MgO is commonly paired with CaCO₃ in the industry for remineralization and pH correction, the impact of equilibrium dynamics in a mixed system is of particular interest and should be investigated in greater detail regarding effluent stability.

In conclusion, the use of MgO is not novel to the water treatment industry; however, this review aimed to provide a synthesis of its uses and potential specific to the drinking water domain. The main drawbacks relate to the need to (i) routinely replenish the media and (ii) use a packed bed contactor that is more expensive than an online chemical injection system. However, it becomes clear that the advantages of using MgO are numerous due to its:

- Quick hydration and the low solubility of Mg(OH)₂, which provides a long-term source of alkalinity and Mg²⁺;
- Higher solubility than calcite;
- pH buffering range and ability to retain divalent metal contaminants as compact hydroxide and oxide precipitates;
- Health benefits associated with magnesium;
- Simpler operation than an alternative online chemical injection system (such as caustic).

It appears, however, that very little work has been conducted to investigate the use of MgO as a simultaneous treatment for remineralization and metal removal, an application that has recently been put forward for CaCO₃ contactors. The potential of the media in this regard is significant and warrants further exploration.

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