Removal of Water Hardness: Comparative Study

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Abstract

Water hardness, resulting from the presence of divalent calcium and magnesium ions, is an important parameter in water treatment due to its effects on household and industrial installations, as well as its broader environmental and health implications. This study presents a comparative evaluation of four hardness-removal techniques: thermal softening, lime–soda treatment, ion exchange using a commercial softening filter, and adsorption using coconut-based granular activated carbon (AC). Artificial hard water with a total hardness of 320 mg $CaCO_3$/L was prepared and analyzed following Polish and ISO standards.Thermal softening removed 39% of total hardness, confirming its selectivity for temporary hardness. The lime–soda process significantly improved hardness reduction, achieving 72% removal under cold conditions and 83% under hot conditions, with enhanced precipitation of $CaCO_3$ and $Mg(OH)_2$ at elevated temperatures. Ion exchange produced the most effective softening, reducing total hardness by 86%, including 83% calcium and 91% magnesium hardness removal. Adsorption studies showed that hardness removal increased with contact time and AC dose, reaching equilibrium after approximately 11 hours and achieving up to 75.5% removal at 12 hours.

The comparative analysis demonstrates substantial performance differences between methods and confirms ion exchange as the most efficient and universal technique. However, adsorption and lime–soda softening provide viable alternatives in contexts where chemical availability, energy consumption, or waste generation are important considerations. These results support informed selection of softening technologies in drinking water treatment and industrial applications.

Keywords: *water hardness, hardness removal techniques, ion exchange, lime-soda softening, granular activated carbon adsorption, comparative analysis*

1. INTRODUCTION

Water hardness – defined as the combined concentration of calcium (Ca^{2+}) and magnesium (Mg^{2+}) ions – remains one of the central challenges in water treatment engineering. It originates primarily from the dissolution of sedimentary rocks, notably limestone and dolomite, into groundwater and surface water systems (Ingin et al., 2024). According to the World Health Organization (WHO), hardness itself is not considered a direct toxicological threat; however, its health-related and technical implications are well documented and widely discussed in the scientific literature (WHO, 2011).

From a human health perspective, both deficiencies and excesses of calcium and magnesium may lead to physiological consequences. Sengupta (2013) notes that excessive calcium intake may promote kidney stone formation, whereas elevated magnesium concentrations can cause gastrointestinal disturbances, including laxative effects. Conversely, epidemiological studies suggest potential benefits of moderate water hardness, including a possible protective effect against cardiovascular disease – an observation highlighted in WHO assessments and several public health reviews (WHO, 2011; Sengupta, 2013; Catling et al., 2008; Huang et al., 2019).

The effects of hardness are particularly pronounced in household and industrial systems. As demonstrated by Hettiarachchi et al. (2017), precipitation of calcium carbonate ($CaCO_3$) and magnesium hydroxide ($Mg(OH)_2$) leads to scale accumulation, reduced pipe diameter, lower heat-transfer efficiency, and increased energy consumption. In controlled boiling experiments, Ingin et al. (2024) showed that calcium ions dominate scale formation, whereas magnesium precipitates more slowly and under more specific pH conditions. These processes contribute to equipment malfunction and rising operational costs, reinforcing the need for effective softening technologies.

To mitigate these issues, numerous methods have been developed for reducing water hardness. Traditional chemical precipitation using lime or soda is widely applied, although it generates substantial sludge volumes – a drawback emphasized in WHO guidelines (WHO, 2011). Modern systems increasingly rely on ion exchange, in which Ca^{2+} and Mg^{2+} ions are replaced with Na^+ or K^+. As highlighted by El-Nahas et al. (2020), ion exchange offers high selectivity and efficiency, but requires periodic resin regeneration and contributes to brine pollution in wastewater.

Growing attention has been directed toward adsorption using natural or waste-derived materials, in line with circular-economy principles (Sepehr et al., 2013; Gupta et al., 2009; Kharel et al., 2016; Lestari et al., 2018; Bekri-Abbesa et al., 2008). Activated agricultural wastes – such as coconut shells, plant biomass, shell waste, or pumice – have shown promising removal capacities. For example, activated carbon derived from coconut shells achieved up to 60% removal of hardness ions under optimized conditions, fitting both Langmuir and Freundlich isotherm models (Bharadwaj et al., 2016; Rolence et al., 2014; Nethaji et al., 2013). Karlsson Faudot (2021) similarly evaluated biomass-based adsorbents, noting their potential but also the challenges associated with scaling them for industrial application.

Membrane technologies, including nanofiltration (NF) and reverse osmosis (RO), provide highly efficient removal of

Tab. 1. Physical parameters of water samples

Tab. 1. Parametry fizyczne próbek wody

Parametr	Artificial hard water	Water after softening
pH	9,23	9,64
Total hardness as $mgCaCO_3/L$	320	195
Calcium hardness as $mgCaCO_3$ /L	180	82
Magnesium hardness as $mgCaCO_3$ /L	140	113

Tab. 2. Physical parameters of water samples

Tab. 2. Parametry fizyczne próbek wody

Parametr	Artificial hard water	Water after cold lime softening	Water after hot lime softening
pH	9,23	9,71	9,33
Total hardness as $mgCaCO_3/L$	320	90	55
Calcium hardness as $mgCaCO_3$ /L	180	60	40
Magnesium hardness as $mgCaCO_3$ /L	140	30	10

Tab. 3. Physical parameters of water samples

Tab. 3. Parametry fizyczne próbek wody

Parametr	Artificial hard water	Water after filtration
pH	9,23	8,1
Total hardness as $mgCaCO_3/L$	320	44
Calcium hardness as $mgCaCO_3$ /L	180	31
Magnesium hardness as $mgCaCO_3$ /L	140	13

divalent ions. As El-Nahas et al. (2020) reported, more than 80% of countries implementing desalination processes use membrane-based softening technologies. Nevertheless, their high energy demand and capital costs limit widespread application. Electrochemical methods, such as electrodialysis and electrocoagulation, represent another promising approach. Helmy et al. (2017) demonstrated that electrocoagulation could remove up to 100% of Mg^{2+} and over 80% of Ca^{2+}, with performance influenced by current density, pH, and mixing conditions.

More recently, innovative materials such as geopolymer–zeolite monoliths have gained attention. Campanile et al. (2022) reported synergistic removal of hardness ions using a geopolymer matrix functionalized with zeolite A and X, achieving 90% Ca^{2+} and 54% Mg^{2+} removal through combined ion exchange and alkaline precipitation mechanisms.

Given the diversity of mechanisms, performance characteristics, and environmental implications associated with different softening technologies, the study of water hardness removal remains a dynamic and evolving research field. The aim of this article is to discuss and compare major softening methods—from conventional to advanced and sustainability-oriented approaches – evaluating their efficiency, advantages, and limitations in removing hardness from water.

2. MATERIALS AND METHODS

2.1. Materials

All chemicals used in this study were of analytical grade and purchased from Chempur. A pH 10 buffer, hardness indicator, and 0.01 M standard EDTA (ethylenediaminetetraacetic acid) were used for water hardness analysis.

Coconut-based activated carbon (AC) was used in the adsorption tests. The adsorbent was in the form of granular activated carbon (GAC), characterized by small and uniform pores, which makes it suitable for the removal of low-molecular-weight contaminants.

A commercially available water softening filter with a cartridge designed for enhanced hardness removal was also used. The main components of the filter media were ion exchange resin and activated carbon.

2.2. Methods

2.2.1 Preparation of artificial hard water

Artificial hard water was prepared by dissolving 288.0 mg of $CaCl_2$, 220.0 mg of $MgSO_4 \cdot 7H_2O$, and 390.0 mg of $NaHCO_3$ in 1L of deionized water. The solution was stored in a plastic bottle (Hettiarachchi et al., 2017).

Water hardness was determined according to PN-ISO 6059:1999, PN-ISO 6058:1999, and PN-C-04554:1999 standards. All hardness values were expressed as mg $CaCO_3$/L. The pH and electrical conductivity of the water were measured using a WTW inoLab® pH 7310 meter. Hardness removal efficiency was calculated using the following equation:

$$removal\ efficiency = (C_i - C_f)/C_i \cdot 100\% \quad (1)$$

Where, C_i and C_f are the initial and final hardness in mg/L respectively.

2.2.2 Water softening using thermal method

To remove temporary hardness, a 0.9 L sample of artificial hard water was boiled for one hour and then cooled to room temperature. The boiling and cooling cycle was repeated two additional times. The sample was subsequently filtered, and the hardness of the filtrate was determined using the EDTA titration method

2.2.3 Water softening using the lime-soda method

Based on the analysis of the artificial hard water, the required doses of lime and soda were calculated using the following equations:

$$Z_1 = 28\ (T_w + T_{Mg} + CO_2 + 0,5) \quad (2)$$

$$Z_2 = 53\ (T_n + 1) \quad (3)$$

where: Z_1 – theoretical dose of calcium oxide (II) [mg CaO/L], Z_2 – theoretical dose of sodium carbonate (I) [mg Na_2CO_3 /L], T_w – carbonate hardness of water before softening [mval/L], T_n – non-carbonate hardness of water before softening [mval/L], T_{Mg} – magnesium hardness of water before softening [mval/L], CO_2 – free carbon dioxide content in water before softening [mval/L].

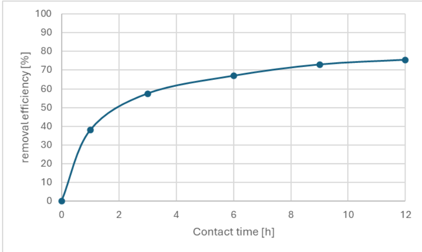

Fig. 1. Effect of contact time on hardness removal by AC (AC quantity = 15g, shaking speed 150 rpm, T=25°C)
Rys. 1. Wpływ czasu kontaktu na usuwanie twardości za pomocą AC (ilość AC = 15 g, prędkość wytrząsania 150 obr./min, T=25°C)

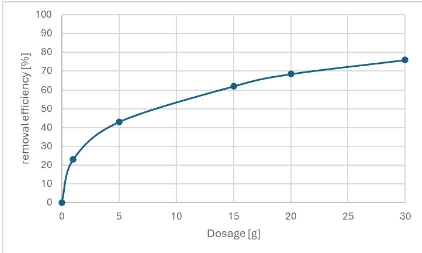

Fig. 2. Effect of adsorbent dose on hardness removal by AC (t=4h, shaking speed 150 rpm, T=25°C)
Rys. 2. Wpływ dawki adsorbentu na redukcję twardości metodą AC (t=4h, prędkość wytrząsania 150 obr./min, T=25°C)

Two water samples of $1dm^3$ each were prepared. The calculated reagent amounts, in the form of lime milk and soda solutions, were added to each sample. The solutions were mixed for two minutes. One sample was kept at room temperature, while the second was heated to 90°C. After softening, the clarified water was filtered, and hardness was determined by titration.

2.2.4 Hardness removal using the softening filter

The filter bed from the cartridge was placed in a glass column with an internal diameter of 5 cm. Artificial hard water was then pumped through the column using a peristaltic pump at a flow rate of 1L/min, corresponding to the filtration speed of a household filter jug. The filtrate was collected, and its hardness was determined using the EDTA titration method.

2.2.5 Hardness removal using activated carbon (AC)

Batch experiments were conducted to optimize contact time and adsorbent dosage.

2.2.5.1. Contact time optimization

In each test, 100 mL of artificial hard water was contacted with 15 g of AC in 300 mL Erlenmeyer flasks. The flasks were shaken in an orbital shaker at 150 rpm and 25°C for predetermined time intervals (1, 3, 6, 9, and 12 hours). After each contact time, the suspension was filtered, and the filtrate was analyzed using the EDTA titration method.

2.2.5.2. Mass optimization

To evaluate the effect of adsorbent dosage, AC samples weighing 1, 5, 15, 20, and 30 g were placed in 300 mL Erlenmeyer flasks. Each sample received 100 mL of artificial hard water. The flasks were shaken for 4 hours at 150 rpm. After the contact time, suspensions were filtered, and the filtrates were analyzed using the EDTA titration method.

3. RESULTS AND DISCUSSION

3.1 Thermal method

The test results are presented in Table 1.

The thermal method involves heating hard water. Under elevated temperature, calcium and magnesium bicarbonates decompose and precipitate at the bottom of the vessel as poorly soluble compounds, according to the following reactions (Surgiel & Kurbiel, 2001; Granops & Kaleta, 2005):

$$Ca(HCO_3)_2 \rightarrow CaCO_3\downarrow + CO_2\uparrow + H_2O \quad (4)$$

$$Mg(HCO_3)_2 \rightarrow MgCO_3\downarrow + CO_2\uparrow + H_2O \quad (5)$$

Magnesium carbonate then undergoes hydrolysis, forming magnesium hydroxide:

$$MgCO_3\downarrow + H_2O \rightarrow Mg(OH)_2\downarrow + CO_2 \quad (6)$$

The results indicate that the thermal method reduces total hardness by 39%, with calcium and magnesium hardness decreasing by 54% and 19%, respectively. These findings confirm that the thermal method removes only carbonate (temporary) hardness, while magnesium remains the primary contributor to permanent hardness. This is consistent with the observations of other researchers (Hettiarachchi et al., 2017).

3.2 Water softening using the lime-soda method

The test results are presented in Table 2.

This method enables the removal of carbonate hardness as well as non-carbonate hardness and dissolved CO_2. Calcium hydroxide reacts with carbon dioxide, leading to the precipitation of calcium carbonate and magnesium hydroxide, and converts non-carbonate magnesium hardness into non-carbonate calcium hardness according to the following reactions (Surgiel & Kurbiel, 2001; Granops & Kaleta, 2005; Szczykowska & Siemieniuk, 2020):

$$CO_2 + Ca(OH)_2 \rightarrow CaCO_3\downarrow + H_2O \quad (7)$$

$$Ca(HCO_3)_2 + Ca(OH)_2 \rightarrow 2CaCO_3\downarrow + 2H_2O \quad (8)$$

$$Mg(HCO_3)_2 + 2Ca(OH)_2 \rightarrow CaCO_3\downarrow + MgCO_3 + 2H_2O \quad (9)$$

$$MgCO_3 + H_2O \rightarrow Mg(OH)_2\downarrow + CO_2 \quad (10)$$

$$MgSO_4 + Ca(OH)_2 \rightarrow Mg(OH)_2\downarrow + CaSO_4 \quad (11)$$

$$MgCl_2 + Ca(OH)_2 \rightarrow Mg(OH)_2\downarrow + CaCl_2 \quad (12)$$

Lime removes carbonate hardness, whereas soda ash eliminates non-carbonate hardness. Sodium carbonate reacts with calcium and magnesium salts according to the following reactions (Surgiel & Kurbiel, 2001; Granops & Kaleta, 2005; Szczykowska & Siemieniuk, 2020):

$$CaSO_4 + Na_2CO_3 \rightarrow CaCO_3\downarrow + Na_2SO_4 \quad (13)$$

$$CaCl_2 + Na_2CO_3 \rightarrow CaCO_3\downarrow + 2NaCl \quad (14)$$

$$MgCl_2 + Na_2CO_3 \rightarrow MgCO_3\downarrow + 2NaCl \quad (15)$$

$$MgSO_4 + Na_2CO_3 \rightarrow MgCO_3\downarrow + Na_2SO_4 \quad (16)$$

Softening using lime–soda does not produce completely hardness-free water due to the limited solubility of $CaCO_3$ and $Mg(OH)_2$. Since the solubility of calcium and magnesium decreases with increasing temperature, warm-lime softening is more effective than cold-lime softening. This is supported by the results showing that cold-lime softening reduces total, calcium, and magnesium hardness by 72%, 67%, and 79%, respectively, whereas warm-lime softening achieves reductions of 83%, 78%, and 93%.

It should also be emphasized that not all salts are removable using this technology. Bicarbonate salts are precipitated by lime, while non-carbonate hardness is converted into soluble sodium salts through reactions with lime and sodium carbonate. Because sodium salts remain dissolved, the lime–soda process cannot eliminate all dissolved solids. Complete removal of such salts requires alternative technologies, such as membrane processes (Zadghaffari, 2013).

3.3. Hardness removal using the softening filter

The test results are presented in Table 3.

Studies have shown that the use of the softening filter reduces total hardness by 86%, while calcium and magnesium hardness decrease by 83% and 91%, respectively. These results confirm that the ion-exchange resin method effectively removes all types of hardness and is the most efficient softening technique.

3.4 Hardness removal using activated carbon (AC)

3.4.1 Contact time optimization

Figure 1 presents the relationship between contact time and hardness removal efficiency.

The hardness removal efficiency increases progressively, reaching 75.5% after 12 hours of contact. As shown in Figure 1, removal efficiency improves with time; however, taking into account both the economic and operational aspects, an optimal contact time of 4 hours can be identified.

The results also indicate that hardness removal reaches equilibrium after approximately 11 hours. Further extension of the contact time does not significantly improve removal efficiency. This behavior may be attributed to the fact that during the initial stages, numerous active surface sites are available for adsorption. Over time, repulsive forces between adsorbed ions and ions remaining in solution hinder further occupation of the remaining active sites (Rolence et al., 2014b).

3.4.2 Mass optimization

Figure 2 presents the influence of adsorbent dosage on hardness removal efficiency at 25°C.

The results show that hardness removal increases with higher AC dosage. A dose of 20 g achieved a removal efficiency of 69%. However, further increases in adsorbent mass resulted in only marginal improvements. This plateau effect is due to the increased availability of exchange and adsorption sites at higher adsorbent concentrations. Once a critical adsorbent dosage is reached, maximum adsorption occurs, and additional AC does not significantly enhance ion removal (Rolence et al., 2014a; Chakrabarty & Sarma, 2012).

4. CONCLUSIONS

Thermal softening effectively removed only temporary hardness, reducing total hardness by 39%. Calcium hardness decreased significantly, while magnesium hardness showed limited reduction, confirming the method's selectivity and its practical limitations for comprehensive softening.

The lime–soda method demonstrated substantially greater efficiency, particularly under hot conditions. Hot lime softening removed 83% of total hardness, outperforming cold lime softening (72%). Elevated temperature improved precipitation of $CaCO_3$ and $Mg(OH)_2$, facilitating removal of both carbonate and non-carbonate hardness.

Ion exchange using a commercial softening filter proved to be the most effective technology evaluated. Total hardness removal reached 86%, with very high reductions in both calcium (83%) and magnesium (91%) concentrations. This confirms ion exchange as the most robust and universal method for complete water softening.

Activated carbon adsorption exhibited moderate hardness removal capacity. Hardness reduction increased with contact time and adsorbent dose, reaching a maximum of approximately 75% after 12 hours. Adsorption equilibrium occurred at around 11 hours. Although less efficient than ion exchange, AC represents a low-cost and sustainable alternative, particularly when waste-derived carbons are used.

The comparative results indicate significant variation in the effectiveness of different softening methods. While ion exchange provides the highest removal efficiency, lime–soda softening and adsorption may be suitable depending on economic constraints, treatment goals, or sludge-generation considerations. Overall, selecting an appropriate softening process should consider not only removal efficiency but also operating conditions, energy requirements, environmental impact, and desired water quality outcomes. The findings of this study provide a comprehensive basis for choosing suitable technologies for drinking water treatment and industrial water management.

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Usuwanie twardości wody: badanie porównawcze

Twardość wody, wynikająca z obecności dwuwartościowych jonów wapnia i magnezu, jest ważnym parametrem w uzdatnianiu wody ze względu na jej wpływ na instalacje domowe i przemysłowe, a także szersze implikacje środowiskowe i zdrowotne. Niniejsze badanie przedstawia ocenę porównawczą czterech technik usuwania twardości: zmiękczania termicznego, obróbki wapnem i sodą, wymiany jonowej z wykorzystaniem komercyjnego filtra zmiękczającego oraz adsorpcji z wykorzystaniem granulowanego węgla aktywnego (AC) na bazie kokosa. Przygotowano i przeanalizowano sztuczną twardość wody o twardości całkowitej 320 mg $CaCO_3$/l zgodnie z normami polskimi i ISO. Zmiękczanie termiczne usunęło 39% twardości całkowitej, co potwierdza jego selektywność w zakresie twardości przemijającej. Proces zmiękczania wapnem i sodą znacząco poprawił redukcję twardości, osiągając 72% usunięcia w warunkach zimnych i 83% w warunkach gorących, ze zwiększonym wytrącaniem $CaCO_3$ i $Mg(OH)_2$ w podwyższonych temperaturach. Wymiana jonowa zapewniła najskuteczniejsze zmiękczanie, redukując twardość całkowitą o 86%, w tym 83% wapnia i 91% magnezu. Badania adsorpcji wykazały, że stopień usunięcia twardości wzrastał wraz z czasem kontaktu i dawką AC, osiągając stan równowagi po około 11 godzinach i osiągając do 75,5% usunięcia po 12 godzinach.

Analiza porównawcza wykazuje znaczne różnice w wydajności między metodami i potwierdza, że wymiana jonowa jest najskuteczniejszą i najbardziej uniwersalną techniką. Jednakże adsorpcja i zmiękczanie wapnem i sodą stanowią realną alternatywę w sytuacjach, w których dostępność chemikaliów, zużycie energii lub wytwarzanie odpadów są istotnymi czynnikami. Wyniki te wspierają świadomy wybór technologii zmiękczania w uzdatnianiu wody pitnej i zastosowaniach przemysłowych.

Słowa kluczowe: *twardość wody, techniki usuwania twardości, wymiana jonowa, zmiękczanie wapnem i sodą, adsorpcja na węglu aktywnym granulowanym, analiza porównawcza*