

# Using Microwave Energy for the Removal of Hardness from Groundwater: Continuous Flow Lab-Scale System

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*Received August 24, 2019; revised and accepted May 29, 2020*

**Abstract:** The present study investigates the characteristics of calcium hardness removal from water using a continuous flow microwave (MW) radiation system with heat exchange. The effects of initial calcium concentration, detention time, and initial temperature were investigated by the study. About 97% of calcium hardness removal was achieved at a detention time of 12.5 minutes. It was concluded from the experiments performed that the optimum conditions for this system use a detention time of 12.5 minutes. This leads to an initial temperature of 70°C when using the heat exchanger. These conditions are valid for the range of Ca initial concentrations between 92 and 204 mg/L as CaCO<sub>3</sub>. The residual concentrations under optimum conditions were 2.4, 2.5, 2.6, and 3 mg/L as CaCO<sub>3</sub> for initial concentrations of 92, 141, 172, and 204 mg/L as CaCO<sub>3</sub>, respectively. The developed system proved to be practical in the continuous flow mode that simulates the actual operations in water treatment plants. It was concluded that MW energy could be one of the most effective methods for large scale removal of hardness from water.

**Keywords:** Hardness, microwave radiation, heat exchange, calcium.

## Introduction

One of the commonest water quality problems around the world is the hardness of water. Globally, only 2.5% of the available freshwater is accessible for human consumption whereas less than 1% of this can be used for drinking purposes (IAEA, 2011). In general, various cations, such as calcium (Ca<sup>2+</sup>) and magnesium (Mg<sup>2+</sup>), present in groundwater are the main cause of hardness in water, as these minerals are picked up from rocks and soils. The hardness by ions concentrated beyond the solubility limit can result in scale deposition.

The scaling problem can lead to plugging in water pipelines and even shut-down water supply systems (Koo et al., 2011). Besides this, water hardness has several adverse impacts on the ability of water usage, either for domestic or industrial applications. Hard water interferes with laundering, washing and bathing. Scaling problems in water pipes, fittings and boilers are also attributed to the hardness due to water (Wu et al., 2013). Moreover, the hard water will inhibit the cleaning efficiency of soap and detergents as the soap tends to combine with minerals existing in water to form a sticky sludge. In addition, some cardiovascular diseases

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such as Alzheimer's and atopic eczema were linked to the frequent utilisation of hard water for domestic purposes (Sengupta, 2013). Therefore, efficient removal of hardness from drinking water is significant to avoid the aforementioned impacts.

Hardness in natural water is mainly caused by Ca and Mg cations and, to less extent, aluminium and iron. Hardness caused due to the number of Ca and Mg cations associated with carbonate ( $\text{CO}_3^{2-}$ ), hydrocarbonate ( $\text{HCO}_3^-$ ) and hydroxide ( $\text{OH}^-$ ) anions are known as carbonate hardness. Typically, water hardness can be removed by using lime ( $\text{CaOH}$ ); also, several treatment processes have been introduced to remove or reduce the hardness from hard waters, such as ion exchange (Janson et al., 2018), reverse osmosis (Min Kyung and Choon, 2017), electrodialysis treatment (Zhang and Chen, 2016), and electrocoagulation (Cabiguen et al., 2018). Recently, microwave (MW) energy has become a promising technology for several environmental applications such as remediation of heavy metals contaminated soil (Jou, 2006; Zhang et al., 2011) and treatment of heavy-metals sludge (Wu et al., 2009). Focussing on water and wastewater treatment, MW energy is involved in different applications such as pre-treatment of petroleum refinery wastewater (Qin and Gong, 2014), removal of ammonia nitrogen in wastewater (Lin et al., 2009; Rabah and Darwish, 2013) and pre-treatment of pharmaceutical wastewater (Qi and Li, 2016). In addition, MW technology has been efficiently utilised for water treatment and purification purposes, such as removal of bisphenol A (BPA) in water (Li et al., 2016) and ballast water treatment (Boldor et al., 2008). To the best of our knowledge, the

application of MW energy in water hardness removal was never tested before. Such a reason for lack of research may be attributed to the high energy demand for water heating by MW radiation. Although such a system was not applied previously, it is assumed that a configuration of MW radiation with a heat exchanger could reduce the required energy. Therefore, the present study aims to investigate the characteristics of removing Ca hardness in water using MW radiation, assisted with heat exchanger.

## Materials and Methods

### Experimental Setup

Figure 1 shows a schematic diagram of the experimental setup. A domestic MW oven (700 W, Dura XB2316, UK) was used as the source of MW radiation. Two holes were drilled at its side cover; one of them as inlet and the other as an outlet for the flowing water, with copper pipes inserted to prevent MW emission. Additionally, a third hole was drilled at the top cover of the oven, wherein a tube connected to a condenser was inserted. The water was pumped into a 300 ml glass tube fixed inside the MW oven. To provide a constant flow rate of water, a peristaltic pump (SP311, VELP Scientifica, Italy) with multiple flow rate settings were connected to a small water tank, to pump the water into the MW oven. Two sampling ports were used to draw samples for the required measurements as shown in Figure 1. Besides, a cartridge filter was used to remove calcium carbonate that precipitated after water drained out from the system.

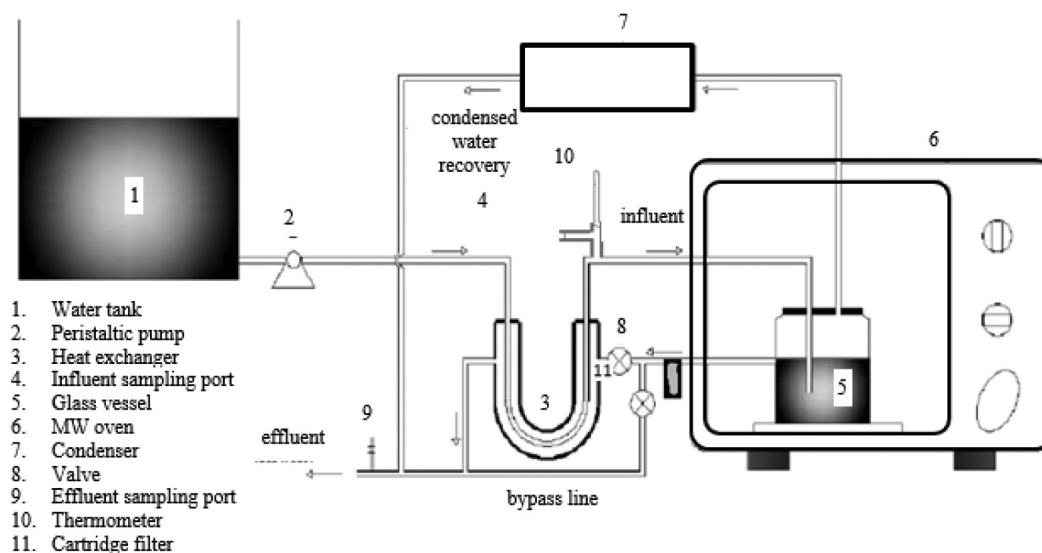


Figure 1: Schematic diagram of the experimental apparatus.

### Heat Exchanger

Heat exchanger is a device built for efficient heat transfer from one fluid to another and is widely used in the engineering processes. This study aims in using heat exchanger to recover effluent heat energy for increasing the influent temperature. The applied heat exchanger consisted of a heat-resistant plastic insulated pipe (inner diameter = 1.95 cm) from which the hot effluent stream passes through it, making a hot water jacket around the cold influent stream pipe (inner diameter = 0.60 cm). It is inserted into the insulated pipe as shown in Figure 2.

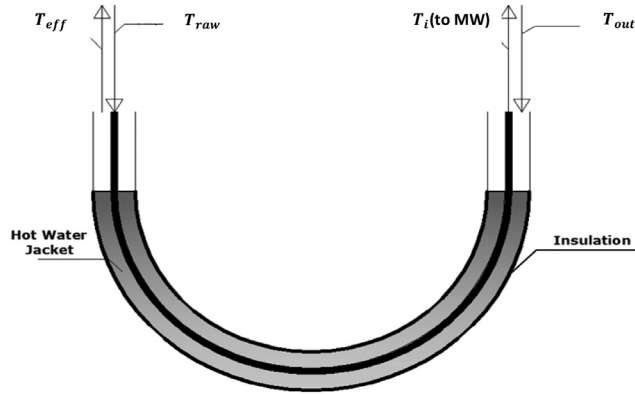


Figure 2: The applied heat exchanger.

Thereafter, the hot effluent ( $T_{out}$ ) entered the heat exchanger with a temperature of 93-95 °C, due to temperature loss in the pipe connecting between the MW oven outlet and the heat exchanger.

To investigate the influence of heat recovery on Ca removal efficiency using a heat exchanger system, the case of 204 mg/L concentration was selected. Four detention times were studied in this experiment: 12.5, 10, 7.5 and 5 min, and five samples were collected at each detention time. In addition, the temperatures of the heated influent ( $T_i$ ) and the cooled effluent ( $T_{eff}$ ) were measured for each flow rate case, using a multi-metre portable device (BoeKel, Model 530, Germany).

### Materials and Analytical Methods

Real groundwater samples were used in this experiment. The samples were obtained from four different wells near Gaza city that had hardness concentrations of 92, 141, 172, and 204 mg/L. The temperature of water samples was measured using a multi-metre portable device (BoeKel, Model 530, Germany). The calcium hardness concentration was measured based on the EDTA standard method. All experiments were run in triplicate and the average value was reported together with its corresponding standard deviation.

### Experimental Procedure

In this research, two sets of experiments were performed; one without the heat exchanger and the other with the heat exchanger. The main objective of applying heat exchanger was to preheat the influent by utilising the temperature of the hot effluent. At the same time, the effluent was cooled to some extent as presented later. The effects of initial calcium concentration ( $C_i$ ), detention time ( $\theta$ ), and initial temperature ( $T_i$ ) were investigated. Four detention times (5, 7.5, 10, and 12.5 min) were tested for each value of  $C_i$  in both experimental sets.

In the second set of experiments (with heat exchanger), one value of  $C_i$  (92 mg/L) was chosen, as this experimental set aims at proving the efficiency of the heat exchanger. In each experimental run, water samples were collected every 5 minutes and the residual Ca concentration ( $C_f$ ) was determined. The system was operated until a steady state of  $C_f$  was achieved.

After heating each sample, the volume of water decreased by about 3-28% of the initial volume due to the evaporation of water. Thus, after collecting each sample, the volume was adjusted with deionised water, to keep the same initial volume of the pumped water. The detention time of water in the MW oven was controlled by altering the pumping flow rate according to the following equation:

$$\theta \text{ (min.)} = \frac{V(\text{ml})}{Q\left(\frac{\text{ml}}{\text{min}}\right)} \quad (1)$$

where  $\theta$  is the detention time in min.,  $V$  is the volume of the glass vessel in ml, and  $Q$  is the water flow rate in ml/min. Table 1 shows the detention time for each flow rate case.

Table 1: Detention time for each flow rate case

Flow rate (ml/min)	Reactor volume (ml)	Detention time (min)	Evaporated water/ detention time period (%)
58	300	5.17	3
40	300	7.5	8
30	300	10	14
24	300	12.5	28

### Results and Discussion

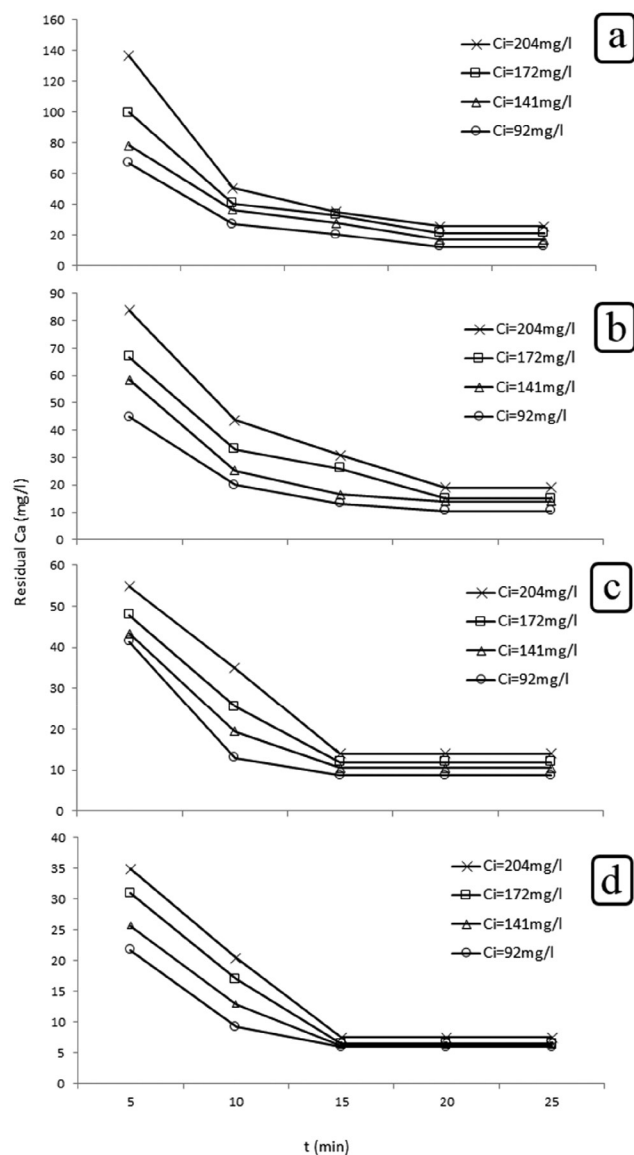
#### Performance of the System

The initial pH of all water samples was found in the range of 7.2-7.5 $\pm$ 0.05, which were not adjusted during this study. With no heat exchanger,  $T_i$  was always 20°C

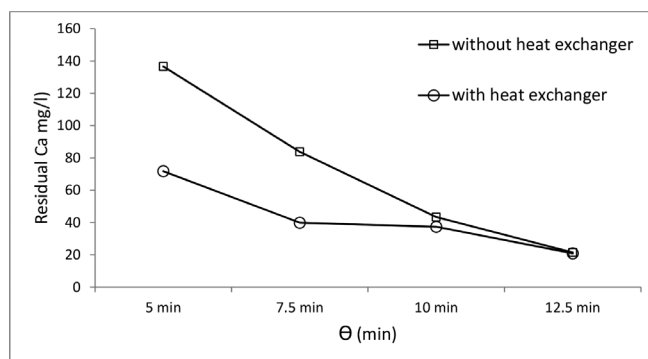
(temperature of the source water) while with the use of heat exchanger,  $T_i$  was raised to 58-70°C.

Figure 3(a-d) shows the variation in  $C_f$  versus  $t$  for different detention time periods. It was observed that  $C_f$  decreased rapidly at the beginning of each experiment and reached a steady-state value after 15 to 20 minutes of heating. This indicates that higher Ca removal was achieved by increasing detention time.

Moreover,  $C_f$  attained in the experiments with heat exchanger was lower than that attained in the experiments without heat exchanger. As shown in Figure 4, when  $\theta$  was 5 min and the  $T_i$  was 20°C, then the  $C_f$  reached 137 mg/L (for  $C_i$  of 204 mg/L) whereas



**Figure 3: Residual Ca concentration versus time without heat exchanger, at (a)  $\theta = 5$  min, (b)  $\theta = 7.5$  min, (c)  $\theta = 10$  min and (d)  $\theta = 12.5$  min.**

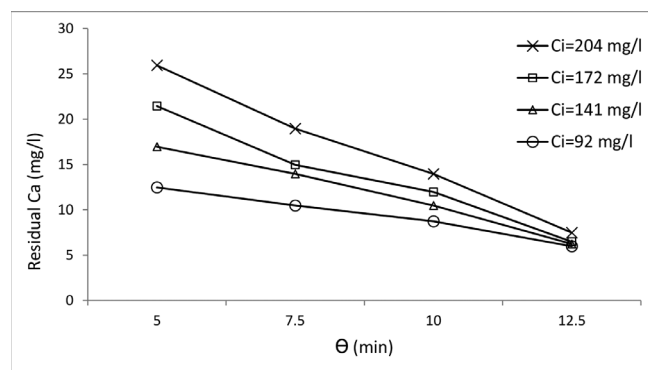


**Figure 4: Effect of using heat exchanger on Ca residual concentration in the 1st sample ( $C_i = 82$  mg/L).**

$C_f$  was reduced to 67 mg/L when  $\theta$  was 5 min and  $T_i$  was 58°C as shown in Figure 6. These results show that better removal efficiency was achieved when using the heat exchanger (33% versus 65% removals).

#### Effect of Detention Time ( $\theta$ )

To investigate the effect of  $\theta$  in each run,  $C_i$  was fixed at 204, 172, 141 and 92 mg/L and  $\theta$  was increased in the range of 5-12.5 minutes. Figure 7 illustrates the effect of detention time (i.e. MW radiation time) on  $C_f$  at steady-state condition, whereas  $T_i$  was kept at 20°C via bypassing the heat exchanger. In general, it was observed that  $C_f$  was decreased significantly with the increase of  $\theta$  inside the MW reactor for all the values of  $C_i$ . For example, for  $C_i$  value of 204 mg/L, the steady state residual Ca concentrations were 26, 19, 14 and 7 mg/L for detention times of 5, 7.5, 10, and 12.5 min, respectively. The same trend was observed for the remaining concentrations as interpreted in Figure 5. This is probably due to the thermal and nonthermal effects of MW radiation. The temperature of water samples could be rapidly increased via the thermal effect that causes intermolecular friction by the alternation in the electromagnetic field. Thus, the rotation of the dipoles



**Figure 5: Effect of detention time on residual calcium concentration at steady state.**



of polar compounds (e.g.  $H_2O$ ) is induced. During the experiments, it was noticed that water samples boiled after approximately 220, 175, 140 and 115 seconds for the flow rates 58, 40, 30 and 24 ml/min, respectively. Therefore, the high temperatures achieved played a vital role in the reaction of  $Ca^{2+}$  ions with bicarbonate ( $HCO_3^-$ ) ions to form insoluble calcium carbonate ( $CaCO_3$ ) as shown in Equation 2:



On the other hand, the fracturing of the intermolecular hydrogen bonds of  $CaHCO_3$  ( $Ca-H \cdots O$  and  $O-H \cdots CaO$ ) could have occurred by inducing a nonthermal effect generated by the frequent vibration of molecular  $CaHCO_3$  and  $H_2O$ , polarised by MW radiation. Therefore, the nonthermal effect of MW radiation could be another valid reason for the precipitation of  $CaCO_3$ .

#### Effect of Initial Calcium Concentration

Four calcium concentrations were tested at this experiment: 204, 172, 141 and 92 mg/L. For each initial concentration, the four flow rate cases were applied, and the optimum concentration was investigated. Figure 6 illustrates the effect of initial calcium concentrations ( $C_i$ ) on the steady-state residual Ca concentration ( $C_r$ ) when the initial temperature ( $T_i$ ) and pH were kept constant at 20°C and 7.5, respectively.

It was observed that at low values of  $\theta$ , the increase of  $C_i$  resulted in a higher concentration of  $C_r$  (i.e., lower removal efficiency), while at high values of  $\theta$  the effect of  $C_i$  on  $C_r$  decreases and becomes negligible. This phenomenon can be attributed to the variations of  $CaCO_3$  formation rate due to the change of the heating energy induced into the water by MW radiation. At low detention time periods, the heating energy is low and has a corresponding low constant  $CaCO_3$  formation. Therefore, when  $C_i$  increases,  $C_r$  also increases. On the

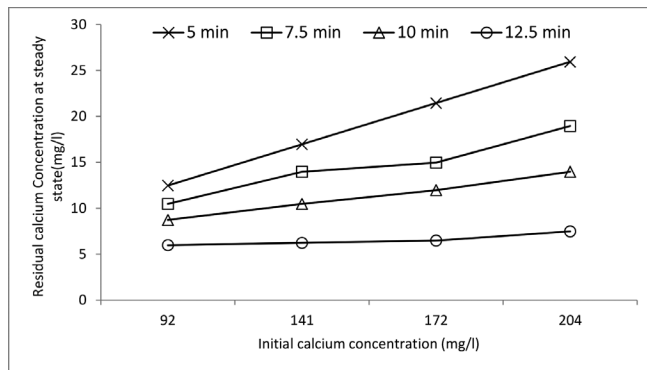


Figure 6: Effect of initial Ca concentration on residual Ca at steady state.

other hand, at high detention time periods, the heating energy is high and has a corresponding high constant formation rate of  $CaCO_3$  that is enough to overcome the increase in  $C_i$  and consequently results in a constant value of  $C_r$  for all values of  $C_i$ .

#### Effect of Initial Temperature

In order to study the effect of initial temperature of water, heat exchanger was used.

It was observed that the temperature of the raw water increased from 21°C to a range between 58–70°C. The maximum heat recovery efficiency of the applied heat exchanger was around 64%, which means that lower detention times and higher flow rates can be applied. Table 2 shows the efficiency of the heat exchanger for each flow rate case.

Table 2: Maximum efficiency of the heat exchanger

Flow rate (ml/min)	Maximum efficiency (%)*
24	64
30	60
40	59
58	52

$$* \text{ Maximum efficiency} = \frac{T_i - T_{raw}}{T_{out} - T_{raw}} \times 100\%$$

Figure 7 shows the relationship between the initial temperature of water and Ca residual. It can be noticed that around 97% of Ca was removed when the initial temperature reached 70°C. Undoubtedly, this result concluded that the heat exchanging approach was able to reduce energy consumption and simultaneously increase the hardness removal efficiency. In this regard, more advanced studies could be conducted to improve this approach and achieve higher efficiencies of heat exchanging.

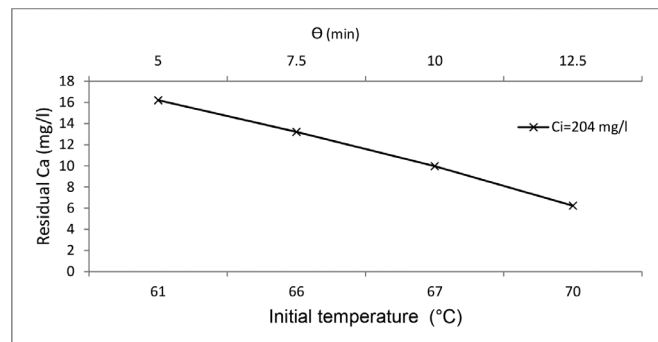


Figure 7: Effect of initial temperature on steady-state Ca residual concentration ( $C_i = 204$  mg/L, with heat exchanger).

## Conclusion

This research was carried out to explore Ca hardness removal from water using a continuous flow MW system equipped with a heat exchanger. The effects of Ca initial concentration, radiation time, and water initial temperature were investigated. It was concluded that the optimum condition of this system is using a detention time of 12.5 minutes that lead to an initial temperature of 70°C when using the heat exchanger. These conditions are valid for the range of initial concentrations between 92 and 204 mg/L as CaCO<sub>3</sub>. The residual calcium concentrations under these conditions were 2.4, 2.5, 2.6, and 3 mg/L as CaCO<sub>3</sub> for initial concentrations of 92, 141, 172, and 204 mg/L as CaCO<sub>3</sub>, respectively. Finally, this study suggests that MW radiation could be a promising technology for the removal of Ca hardness from water.

## Acknowledgement

The authors would like to extend sincere thanks to The Islamic University-Gaza, Universiti Teknologi Malaysia (UTM) and Ministry of Higher Education Malaysia (Fundamental Research Grant Scheme (FRGS) No. 5F249) for the assistance in conducting and publishing this study.

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