

Short Communication

Desalination Behavior of Calcined Hydrotalcite From Seawater for Preparation of Agricultural Cultivation Solution Using Natural Zeolite

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Abstract

The desalination behavior of calcined hydrotalcite (CHT) from seawater used to treat seawater that was subsequently subjected to zeolite treatment prior to application of irrigation water was investigated. The results revealed that CHT removed Cl^- , SO_4^{2-} , Mg^{2+} and Ca^{2+} from seawater, but that Na^+ and K^+ were almost unchanged, and that CHT treatment made the pH of the solution alkaline. As the solution temperature increased, the solution pH increased, the rate of Mg^{2+} , Ca^{2+} , Cl^- and SO_4^{2-} removal improved, and greater Cl^- removal was observed. Following CHT treatment, the obtained solution (CHT-solution) had a high Na^+ content and high pH (approximately 12), while treatment of CHT-solution with natural zeolite neutralized the pH of the CHT-solution and reduced the Na^+ content. Although germination of radish sprouts did not observed using seawater and CHT-solution, the growth of radish sprouts could be observed using the final solution after natural zeolite treatment of CHT-solution.

Keywords: desalination, calcined hydrotalcite, seawater, natural zeolite, radish sprout

1. Introduction

In the 21st century, problems associated with energy shortages and environmental conditions have increased, and food shortage has become one of the greatest problems worldwide. Securing irrigation water for agriculture is one of the most important factors in food production; however, regions subjected to shortages of agricultural water are increasing as a result of desertification and global warming. Indeed, during the 20th century, the global human population increased fourfold, but indispensable amount of water from natural freshwater sources increased eightfold (Abdul, 2006).

Effective utilization of untapped resources is an important issue in the twenty-first century (World Water Council [WWC], 2002). One method of the technology for supplying water resources is seawater desalination. The multi-stage flash (MSF) and the reverse osmosis membrane (RO) are well-known desalination technologies (Goto, 2000) in which almost all ions contained in seawater are removed, producing high-quality fresh water for domestic or industrial use; however, 70% of global fresh water use is used for irrigation (Tilzer, 2006). Irrigation water for agriculture use should contain elements for crops rather than being highly purified, and use of MSF or RO for agriculture is not cost effective. High-purity water requires the fertilizer addition before application to the agricultural use. Seawater contains essential elements needed for plant growth, but its high concentration of NaCl causes salt damage, which prevent its direct use. Accordingly, irrigation water produced from seawater by reducing NaCl content using a simple process would be extremely advantageous.

In an earlier study, we attempted a novel two-step process to reduce the NaCl concentration in seawater using two clays, calcined hydrotalcite (CHT) and natural zeolite, which resulted in generation of a solution capable of supporting the growth of radish sprouts (Wajima et al., 2006; Wajima et al., 2008,2009, 2010). Hydrotalcite is a layered double hydroxide composed of a brucite-like positively charged layer due to the partial substitution of Mg^{2+} and Al^{3+} , which is compensated for by negatively charged interlayers containing anions and water (Constantino & Pinnavaia, 1995; Rives & Ulibarri, 1999; Pavan et al., 1999). Hydrotalcite calcined at 500–800 °C is transformed into Mg-Al oxide, which can adsorb anions from aqueous solutions through the

reconstruction of their original structure (Pavlovic et al., 2005). Anion contaminants removed from the solution by CHT have been reported and shown to have high removal capacity for acids (Vreysen & Maes, 2008; Yang, Song, & Qian, 2010, 2013), heavy metal ions (Goswamee et al., 1998; Yang et al., 2005), pesticides (Inachio et al., 2001; You, Zhao, & Vance, 2002; Legrouri et al., 2005; Li et al., 2005; Cardoso & Valim, 2006), dyes (Zhu et al., 2005; Gaini et al., 2009; Asouhidou et al., 2011), halogens (Lv et al., 2006; Lv & Luo, 2007), phosphates (Shin et al., 1996; Chitrakar et al., 2005; Das et al., 2006; Cheng et al., 2009), surfactants (Pavan, Crepaldi, & Valim, 2000) and thiocyanate (Li et al., 2008), but there have been no studies of the treatment of seawater using CHT. Natural zeolite is generally associated with grassy volcanic rock in natural deposits, and is available in large quantities at low cost (Tominaga, 2001). Due to the ion-exchange capacity, absorption characteristics, water retention and low cost, natural zeolites have been used in various fields, such as agronomy, horticulture and industry (Akgü et al., 2006). Natural zeolite contains exchangeable cations, such as Mg^{2+} , Ca^{2+} , K^+ and Na^+ . When seawater is treated with natural zeolite, Na^+ in seawater is exchanged with exchangeable cations in zeolite, and Na^+ can be removed from seawater and supply Mg^{2+} , Ca^{2+} and K^+ to the solution (Wajima, 2013). This process can be applied to seawater after its use as a coolant in electric power plants and chemical plants, and to high salinity water in some areas; accordingly, we expect this process to be useful for vegetation in arid areas. Therefore, we investigated the ion exchange properties of natural zeolite in a two-step process (Shimizu et al., 2007; Wajima et al., 2013). However, little information regarding the treatment of seawater using CHT is available.

In this study, we examined the desalination properties of calcined hydrotalcite from seawater to determine the conditions for anion reduction. In addition, solutions obtained via a combined process of calcined hydrotalcite and natural zeolite were tested for the growth of radish sprouts to examine their potential for use as irrigation water.

2. Experimental

2.1 Samples

Two types of Japanese seawater were used in this study. Seawater-1, which was used for the CHT treatment experiment, was obtained from the surface of Imari Bay, Saga Prefecture, Japan. Seawater-2, which was used for the cultivation test, was collected from other points below 100 m depth in Japan. The chemical compositions and pH values of seawater used in this study are shown in Table 1. The properties of the seawater used were similar and typical of seawater found worldwide (Nakashima, 2002).

Table 1. Chemical compositions and pH of seawater used in this study

	Chemical composition (mg/L)						pH
	Na^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	SO_4^{2-}	
Seawater-1	10,753	361	1256	378	16715	2476	7.4
Seawater-2	11,283	412	1382	334	16370	2823	8.0

Commercial CHT, KW-2000, which was supplied from Kyowa Kagaku Kogyo Company Limited, Japan and Japanese natural zeolite, Iizaka-zeolite obtained from Fukushima prefecture were used. The exchangeable cations and cation exchange capacity (CEC) of natural zeolite are shown in Table 2. Natural zeolite samples had high CEC values, and most of the exchangeable cation sites were occupied by Ca^{2+} and Na^+ ions.

Table 2. Exchangeable cations and cation exchange capacity of natural zeolite used in this study

Amount (mmol/g)	Exchangeable cations				Cation exchange capacity
	Na^+	K^+	Mg^{2+}	Ca^{2+}	
	0.56	0.21	0.03	0.44	1.67

2.2 Removal Behavior of Calcined Hydrotalcite

We examined the effects of reaction temperature and reaction time on the pH of the solution and levels of Na^+ , K^+ , Mg^{2+} , Ca^{2+} , Cl^- and SO_4^{2-} in seawater after CHT treatment.

Briefly, 50 g of CHT were added to 500 mL of seawater and stirred with a magnetic stirrer at 5, 15, 25, 40 and 60 °C. During stirring, 3-mL aliquots of the slurry were removed at varying time intervals to determine the pH of the solution and concentrations of Na^+ , K^+ , Mg^{2+} , Ca^{2+} , Cl^- and SO_4^{2-} in the seawater to monitor the reaction process.

2.3 Combination of CHT Treatment and Natural Zeolite Treatment

We examined the possibility of preparing agricultural cultivation solution using a two-step combination process of CHT treatment and natural zeolite treatment. Briefly, 50 g of CHT were added to 500 mL of seawater-2 and stirred with a magnetic stirrer at 25 °C for 6 h. After stirring, the slurry was filtered to give CHT-solution. Next, 20 g of natural zeolite were added to 200 mL of CHT-solution and stirred for 1 h with a magnetic stirrer. After stirring, the slurry was filtered and fresh natural zeolite was added. This procedure was repeated ten times to obtain the final solution. The pH of the seawater-2, CHT-solution and final solution, and the contents of Na^+ , K^+ , Mg^{2+} , Ca^{2+} , Cl^- and SO_4^{2-} in these solutions were determined, after which they were applied as potential agricultural cultivation solutions.

2.4 Cultivation Test

The solutions obtained during the process were applied to growth test using radish sprouts (*Raphanus sativus*). During this investigation, cultivation was conducted for 10 days at room temperature to investigate the possibility of using the obtained solutions.

2.5 Characterization

The pH of the solution was measured with a pH meter (MA-130, Mettler Toledo). Concentrations of Na^+ , K^+ , Mg^{2+} , Ca^{2+} , Cl^- and SO_4^{2-} in the solution were determined by ion chromatography (DX-120, Japan Daionex). Removal percentages of Cl^- and SO_4^{2-} , R (%), were calculated using the following equation:

$$R = (C_0 - C) / C_0 \times 100 \quad (1)$$

where C_0 and C are the concentrations of Cl^- and SO_4^{2-} in the initial solution and the treated solution (mg/L), respectively.

Exchangeable cations and cation exchange capacities of natural zeolite were measured by the modified Schollenberger's method (Development of New Utilization of Materials [DNUM], 1994).

3. Results and Discussion

3.1 Desalination Properties of Calcined Hydrotalcite

Figure 1 shows the change in pH of the solution at 5, 25 and 60 °C after adding CHT. Before the addition of CHT, the pH of the seawater was 7.4, while after the addition of CHT, the pH of the solutions became highly alkaline. The pH of the solution gradually increased to about 12 as temperature increased from 5 °C to 60 °C.

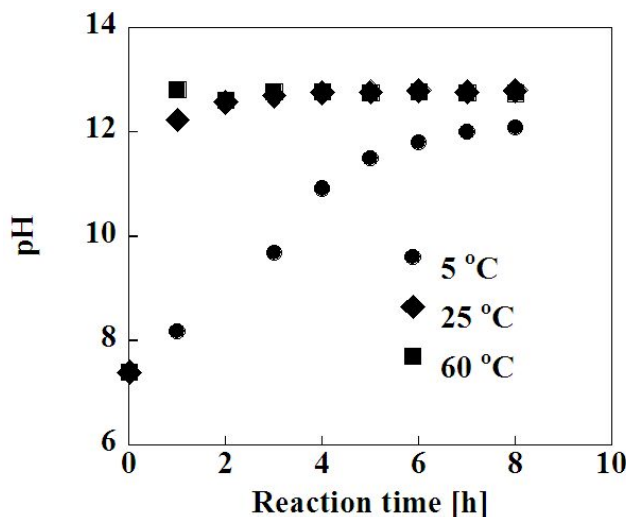


Figure 1. Change in solution pH at 5, 25 and 60 °C after adding calcined hydrotalcite

Figure 2 shows the main cations concentrations in seawater, (a) Na^+ , (b) K^+ , (c) Mg^{2+} and (d) Ca^{2+} , at various temperatures after adding CHT. During stirring, the concentrations of Na^+ and K^+ remained almost constant, regardless of temperature, indicating that their levels were unchanged following treatment with CHT. However, the level of Mg^{2+} and Ca^{2+} decreased slowly at 5 °C, and they reached zero after 1 h at 15 °C. These behaviors correlate well with the behavior of pH (Figure 1), which was likely because increasing pH would cause Mg^{2+} and Ca^{2+} to hydroxylate and precipitate to reduce the contents of Mg^{2+} and Ca^{2+} in seawater (Kragten, 1978).

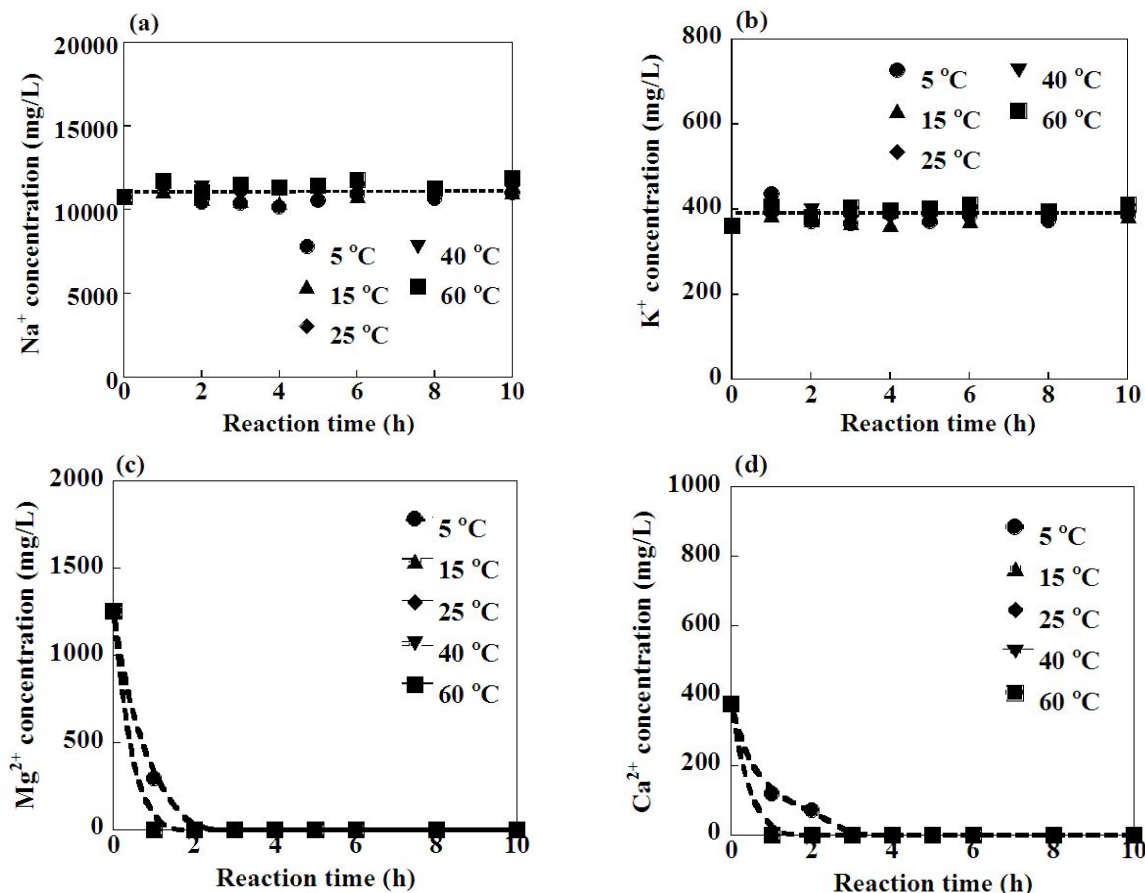


Figure 2. Change in the concentrations of main cations in seawater, (a) Na^+ , (b) K^+ , (c) Mg^{2+} and (d) Ca^{2+} , at various temperatures after the addition of calcined hydrotalcite

Figure 3 shows the changes in the concentrations of the main anions in seawater, (a) Cl^- and (b) SO_4^{2-} at various temperatures after adding CHT. During stirring, the concentrations of Cl^- and SO_4^{2-} decreased and became almost constant. For Cl^- , as temperature increased, the reaction time to equilibrium concentration increased, as did the reduction in Cl^- . It is noted that the removal percentages of Cl^- at 1 h treatment are 4.8% (5 °C), 5.0% (15 °C), 15.2% (25 °C), 28.1% (40 °C) and 34% (60 °C). For SO_4^{2-} , no significant difference of reduction rate was observed at all setting temperatures in this experiment. The enough time to reach equilibrium for the removal of Cl^- and SO_4^{2-} was over 2 h under all conditions.

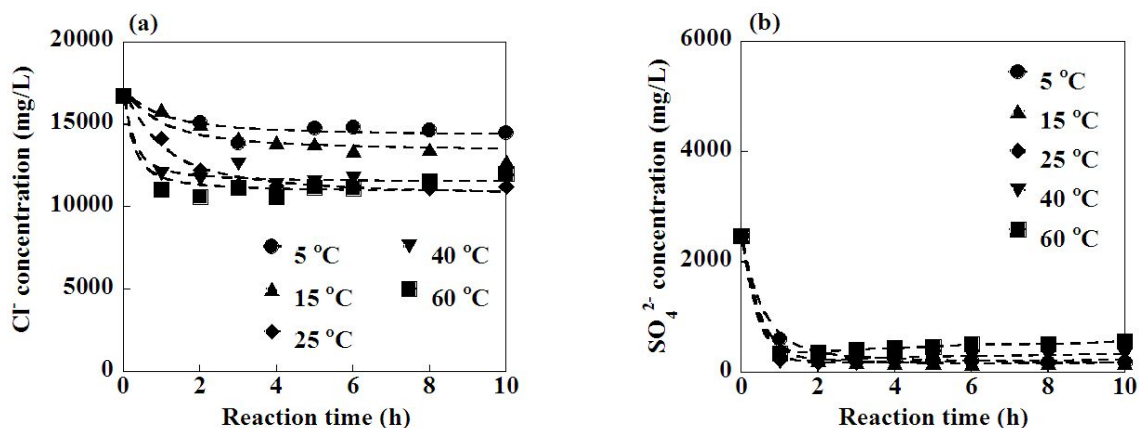


Figure 3. Change in concentrations of main anions in seawater, (a) Cl^- and (b) SO_4^{2-} , at various temperatures after adding calcined hydrotalcite

Figure 4 shows the temperature dependence of the removal of Cl^- and SO_4^{2-} from seawater, and the pH of the solution after 6 h of stirring. Regardless of the temperatures, the removal of SO_4^{2-} remained almost constant (80–90%). As the temperature increased to 25 °C, removal of Cl^- and pH of the solution increased, but above 25 °C, these values remained almost constant.

Based on these results, the condition using CHT at 25 °C over 2 h is enough to remove anions from seawater, is the optimum condition.

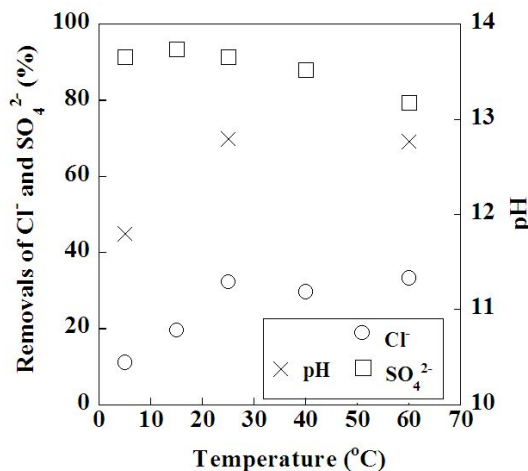


Figure 4. Temperature dependence of Cl^- and SO_4^{2-} removal from seawater and pH of the solution after 6 h of stirring

3.2 Cultivation

Radish sprouts were cultivated with seawater-2, CHT-solution and final solution.

Table 3 shows the chemical compositions and pH of seawater, CHT-solution and final solution. Seawater contained high concentrations of some ions, especially Na^+ (11,283 mg/L) and Cl^- (16,370 mg/L). The concentrations of Cl^- and SO_4^{2-} were highly reduced in CHT-solution, and the pH of solution was high (pH 12.1) because CHT fixes anions and releases OH^- (Kameda, Yoshioka, & Okuwaki, 2001). The concentrations of Mg^{2+} and Ca^{2+} also decreased. In the final solutions, the pH became neutral, Na^+ decreased, and Mg^{2+} and Ca^{2+} (which were lost in the first step) increased because of ion exchange between Mg^{2+} and Ca^{2+} in the natural zeolite and Na^+ in the CHT-solution.

Radish sprouts were grown in each solution for 10 days at room temperature (Figure 5). We could observe the growth of radish sprouts using the product solution through a two-step process (Figure 5 (c)), although we could

not observe the germination of radish sprout using seawater and the CHT-solution (Figure 5 (a), (b)). Solutions prepared from seawater by this process were possible for use as cultivation solutions, and confirm that this process can be applied to seawater collected from various points.

Table 3. Chemical compositions and pH of seawater, Solution-1 and Solution-2

	Chemical composition (mg/L)						pH
	Na ⁺	K ⁺	Mg ²⁺	Ca ²⁺	Cl ⁻	SO ₄ ²⁻	
Seawater-2	11283	412	1382	334	16370	2823	8.0
CHT-solution	13640	504	8	36	13483	584	12.1
Final solution	1291	62	54	1347	8622	433	7.1

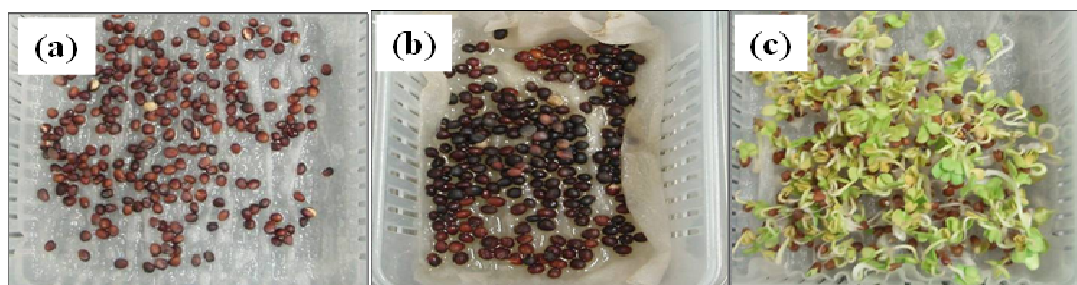


Figure 5. Observation of *Raphanus sativus* after 10-days of growth using (a) seawater, (b) CHT-solution, and (c) final solution

4. Conclusion

A novel process for preparing agricultural cultivation solution from seawater using two clays, hydrotalcite and natural zeolite was attempted. We investigated the desalination properties of water treated using calcined hydrotalcite, and those of water treated by a combination of ion exchange processes using natural zeolite, after which treated water was applied to determine if it could be used to grow Radish sprouts. Treatment with calcined hydrotalcite reduced Mg²⁺, Ca²⁺, Cl⁻ and SO₄²⁻ from seawater and increased the pH to high levels. The optimum treatment conditions using calcined hydrotalcite were 6 hours at 25 °C. Treatment with natural zeolite could neutralize highly alkali solution after calcined hydrotalcite treatment, and reduce Na⁺ and supply Mg²⁺ and Ca²⁺ to the solution via ion exchange with natural zeolite. Radish sprouts could be grown using the solution obtained from treatment with a combination of ion exchange processes using natural zeolite.

From these results, it is possible to prepare agricultural cultivation water from seawater using combined treatment with calcined hydrotalcite and ion exchange using natural zeolite.

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