Regular Article

Characteristics of Raw and Acid-Activated Bentonite and Its Application for Improving Electrical Conductivity of Tap Water

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This study aimed to investigate the characteristics of acid-activated bentonite by focusing on its capability of improving the quality of tap water used during wire electrical discharge machining. Raw bentonite (RB) was activated using sulfuric acid, nitric acid, and phosphoric acid solutions with concentrations of 1, 5, and 10 mol/L, respectively. Scanning electron microscopy images, specific surface area, pore volume, cation exchange capacity, X-ray diffraction patterns, and binding energy of RB and acid-activated bentonites were also evaluated. The specific surface area and pore volume of acid-activated bentonites exceeded those of RB. Conversely, the cation exchange capacity of acid-activated bentonites exhibited an opposite trend. The electrical conductivity of tap water was decreased significantly due to bentonite activated with sulfuric acid, nitric acid, and phosphoric acid solution (removal percentage of approximately 31–39%), as compared to that due to RB. Therefore, the relationship between electrical conductivity and the removed concentration of anion/cation ions was evaluated; the correlation coefficient was -0.950 for the experimental condition in this study. Additionally, the amount of magnesium, calcium, potassium, and sodium ions were decreased after the treatment. These results indicated that acid-activated bentonite can be produced from RB *via* acid activation and that it can be used to decrease electrical conductivity of tap water.

Key words bentonite; acid activation; electrical conductivity; tap water; wire electrical discharge machining

Introduction

Wire electrical discharge machining has been extensively applied in numerous processes, such as those used in aerospace and automotive industries and for constructing surgical components, due to its capability of machining intricate shapes and profiles irrespective of the hardness of the materials.^{1–3} During this process, a thin wire is employed as the electrode to cut the workpiece in a dielectric fluid. Additionally, this process can also be used to easily remove materials of any hardness *via* melting and vaporization.⁴ The performance of wire electrical discharge machining is dependent on four parameters: electrical parameters (*e.g.*, plus on time and plus off time), non-electrical parameters (*e.g.*, dielectric flow rate and dielectric conductivity), workpiece parameters (*e.g.*, material type and size), and wire electrode parameters (*e.g.*, wire speed and wire tension).^{5,6} Several researchers reported improvements in the performance of wire electrical discharge machining by using different modeling and optimization techniques.^{1,7}

During the application of wire electrical discharge machining, tap water is often utilized as a dielectric fluid because machining under tap water leads to a superior material removal rate and lower wear ratio than those when machining in kerosene. Additionally, if kerosene is used in wire electrical discharge machining, it decomposes and produces harmful vapors such as carbon monoxide and methane.^{8,9} In a previous study, optimal machining rates were realized using tap water.⁶ However, when using tap water for wire electrical discharge

machining, electrical discharges can easily occur due to the high resistivity of tap water. This indicates an increase in the ion concentrations released from the wire or workpiece.^{10,11} This phenomenon causes a short and damages the wire or workpiece.¹² Therefore, the electrical conductivity of tap water used in this process is usually controlled *via* an ion exchange resin. However, ion exchange treatments that use ion exchange resins are relatively expensive and not environment-friendly. Thus, alternative approaches are required.

In this study, we focused on clay mineral adsorbents, instead of the conventional ion exchange resins, due to their abundance and low price.^{13,14} Bentonite is a type of natural clay mineral that is abundantly available worldwide.¹⁵ Bentonite is mainly composed of montmorillonite, which belongs to the 2:1-type clay family. Its basic layer structure comprises an octahedral sheet, which includes aluminum placed between two tetrahedral sheets that include silicon.¹⁵ Previous studies reported that bentonite exhibits a large specific surface area, cation exchange capacity, and capability of adsorbing organic and inorganic ions from aquatic media.^{16–18} Additionally, the capability of bentonite in adsorbing metal ions is observed during ion exchange at the permanent-charge sites and during the formation of complexes with surface hydroxyl groups.¹⁹ Furthermore, many researchers reported that acid activation treatment of bentonite increases its specific surface area, mesoporosity, surface acidity, and capability of adsorbing metals.^{20–22} However, there is a paucity of studies on the application of acid-activated bentonite for improving the quality of

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Fig. 1. SEM Images of Adsorbents

Scale bar shows 50 μm . SU1510 microscope is used for measurement with an accelerating voltage of 5 kV.

tap water used in wire electrical discharge machining. For this purpose, it is important to investigate both raw bentonite (RB) and acid-activated bentonite.

This study aims to improve the quality of tap water used for wire electrical discharge machining; the study focuses on decreasing the electrical conductivity of tap water using a low-cost adsorbent such as RB or acid-activated bentonite. This investigation also constituted a fundamental study, which involved the application of acid activated bentonite for the removal of ions from tap water.

Experimental

Materials In the study, RB and the required acid solutions were purchased from FUJIFILM Wako Pure Chemical Corporation (Japan). Acid-activated RB was prepared *via* the following method: Five grams of RB was mixed with 50 mL of sulfuric acid, nitric acid, and phosphoric acid solutions at concentrations of 1, 5, and 10 mol/L, respectively. The reaction mixtures were shaken at 200 rpm for 4 h under ambient conditions. Subsequently, the suspensions were filtered through a 0.45- μm membrane filter (Advantec MFS, Inc., Japan), and the residue was then washed with purified water. The obtained sample was dried at 60 $^{\circ}\text{C}$ for 1–2 d. The samples are labeled as follows: bentonite treated with sulfuric acid at 1 mol/L is denoted as BS1, bentonite treated with nitric acid at 5 mol/L is denoted as BN5, and bentonite treated with phosphoric acid at 10 mol/L is denoted as BP10. In the study, we prepared nine samples, denoted as BS1, BS5, BS10, BN1, BN5, BN10, BP1, BP5, and BP10. Tap water was obtained from a faucet at Kindai University in Higashi-Osaka (Japan).

Characteristics of Adsorbents The morphologies of the prepared adsorbents were measured *via* a scanning electron microscope (SEM, SU1510 microscope, Hitachi High-technologies Co., Japan) with an accelerating voltage of 5 kV. Additionally, X-ray diffraction pattern analyses were performed using MiniFlex II (Rigaku, Japan) with $\text{CuK}\alpha$ and $K\beta$ filter. The broad-range pattern ($2\theta = 5\text{--}90^{\circ}$, with a voltage of 30 kV and current of 15 mA) was collected under ambient conditions. The specific surface area and pore volume were measured

using the NOVA4200e instrument (Quantachrome Instruments Japan G.K., Japan) based on adsorption/desorption isotherms of nitrogen. The cation exchange capacity (CEC) was determined using the Japanese Industrial Standard method (JIS 1478). Additionally, the binding energy was analyzed using the AXIS-NOVA instrument (Shimadzu Co., Ltd., Japan) under an accelerating voltage of 15 kV and electric current of 10 mA. The pH of the solution was measured using the F-73S digital pH meter (HORIBA, Ltd., Japan).

Analysis of Tap Water The electrical conductivity of tap water was measured by using the multi-water quality checker conductivity meter WA-2017SDJ (Sato Shoji, Japan). Concentrations of sodium, magnesium, aluminum, potassium, calcium, and iron ions were measured by means of inductively coupled plasma optical emission spectrometry (ICP-OES, iCAP-7600 spectrometer, Thermo Fisher Scientific Inc., Japan). Additionally, concentrations of chloride, nitrate, and sulfate ions were measured using the ion chromatography instrument DIONEX ICS-900 (Thermo Fisher Scientific Inc.). The measurement conditions reported in a previous study were employed.²³⁾ Standard solutions of sodium ion (NaCl in water), magnesium ion ($\text{Mg}(\text{NO}_3)_2$ in 0.1 mol/L HNO_3), aluminum ion ($\text{Al}(\text{NO}_3)_3$ in 0.5 mol/L HNO_3), potassium ion (KCl in water), calcium ion (CaCO_3 in 0.1 mol/L HNO_3), and iron ion ($\text{Fe}(\text{NO}_3)_3$ in 0.1 mol/L HNO_3) were obtained from FUJIFILM Wako Pure Chemical Corporation. A standard solution of anions (anion mixed standard solution IV) was purchased from Kanto Chemical Co., Inc. (Japan).

Application of Acid-Activated Bentonite First, 0.1 g of each adsorbent was mixed with 100 mL of tap water. Second, the reaction mixture was shaken at 250 rpm for different intervals (0, 0.17, 0.5, 1, 2, 6, 22, and 24 h) under ambient conditions. Finally, the suspensions were filtered through a 0.45- μm membrane filter (Advantec MFS, Inc., Japan). The changes in electrical conductivity, concentration of each solution, and solution pH were measured using previously reported methods. The results of the study are presented as mean $\pm$ standard deviation ($n = 2\text{--}4$, Sections, “Analysis of Tap Water” and “Application of Acid-Activated Bentonite”).

Results and Discussion

Effect of Acid Activation on Characteristics The SEM images of each adsorbent are shown in Fig. 1. No changes were observed in the surface roughness of RB and acid-activated bentonite for the experimental conditions. The characteristics of the adsorbents are summarized in Table 1. The specific surface area and cationic exchange capacity of RB were 32.3 m²/g and 100.1 cmol/kg, respectively. Specifically, the mesopore volume increased significantly compared to the micropore and macropore volumes. Furthermore, increases in the specific surface area and pore volumes were observed after the acid activation of bentonite *via* sulfuric acid, nitric acid, and phosphoric acid solutions at different concentrations. This was due to leaching of the octahedral cations from the interlayer of the adsorbents and the subsequent re-generation of the pores during acid activation treatment. Similar trends were observed in previous studies when using hydrochloric acid.^{24,25)} The values of the specific surface area and pore volume of the BS and BN series exceeded those of the BP series. However, significant differences in the values were not confirmed for each concentration. The specific surface area of BN10 was lower than those of BN1 and BN5, and this is potentially explained by the process of passivation.^{24,26)} Conversely, the CEC of acid-activated bentonite was lower than that of RB for the experimental conditions considered in this study. Similar results were reported in previous studies.^{24,26,27)} The cations in the octahedral sheet were released into the solution due to the acid activation treatment; consequently, the crystal structure was partially destroyed.^{26,27)} Thus, the CEC decreased after the acid activation treatment.

Figure 2 shows the X-ray diffraction (XRD) patterns of RB and acid-activated bentonite. The XRD patterns of the adsorbents were evaluated using the powder diffraction file 2010 (The International Center for Diffraction Data). The RB was composed of montmorillonite Na_{0.3}(Al Mg)₂Si₄O₁₀OH₂·6H₂O, quartz (SiO₂), and opal CT (SiO₂). Opal CT is mineral paracrystalline silica, which does not exhibit an exact crystal structure.²⁸⁾ As reported in a previous study, montmorillonite minerals exhibit two types of XRD patterns, namely basal and non-basal reflections (*hk* bands). The basal and non-basal reflections correspond to the hydration and the interlayer hydration.²⁹⁾ In the study, we observed the *d*(001) peak (the first peak appeared at *2θ* of approximately 7.31°), which indicates that hydronium cations are present in the interlayer of RB and can be exchanged with cations without a degradation in the

layer structure.²⁴⁾ Additionally, we observed *hk* bands at 20.00,

Fig. 2. XRD Patterns of Adsorbents

MiniFlex II is used for measurement with CuKα and Kβ filter. The broad-range pattern *2θ* = 5 to 90 with voltage corresponding to 30 kV, and current corresponding to 15 mA. ▲ Montmorillonite Na_{0.3}(Al Mg)₂Si₄O₁₀OH₂·6H₂O ■ Quartz SiO₂ ● Opal CT SiO₂. (Color figure can be accessed in the online version.)

Table 1. Characteristics of Adsorbents

Adsorbents	Specific surface area (m ² /g)	Pore volume (μL/g)			Cation exchange capacity (cmol/kg)
		<i>d</i> ≤ 2 nm	2 < <i>d</i> ≤ 50 nm	<i>d</i> > 50 nm	
RB	32.3	0.2	72.6	15.4	100.1
BS1	59.3	0.1	93.6	19.1	66.5
BS5	59.0	0.6	96.3	30.3	65.7
BS10	57.9	0.6	85.6	22.5	69.3
BN1	57.8	0.1	100.1	15.7	69.7
BN5	66.9	0.3	114.5	14.2	70.9
BN10	50.4	0.7	92.2	12.8	81.9
BP1	52.9	0.3	72.0	13.9	73.2
BP5	51.7	0.2	84.1	20.5	72.3
BP10	52.7	0.5	83.8	17.7	72.4

35.16, and 61.90° (2 θ).

A broad and weak $d(001)$ peak was observed following the acid activation of bentonite using sulfuric acid, nitric acid, and phosphoric acid, indicating that the crystallinity of RB was significantly affected due to the acid activation treatment. Specifically, the $d(001)$ peak (2 θ) shifted from 7.31° to 6.03–6.44°, 6.38–6.57°, and 6.48–6.97° for the BS, BN, and BP series, respectively. However, this peak did not completely disappear. Furthermore, the changes in non-basal reflections were less than those in basal reflections. A previous study has reported that this change is likely due to the release of cations from the octahedral sheet of the RB structure.^{28,30–32} Similar trends were observed in this study. A few studies reported that an amorphous structure can be obtained after the acid activation treatment of bentonite using sulfuric acid, nitric acid, and phosphoric acid.³³ However, very few studies have evaluated the effects of acid concentrations on the properties of bentonite. Thus, our results differ from those in previous reports, indicating that the relationship between the acid attack and structure preservation was important in terms of preparing acid-activated bentonite. Additionally, significant changes in the XRD patterns were not observed for the acid activation using each acid, thereby supporting the characteristics of adsorbents listed in Table 1.

Subsequently, the binding energies of the surfaces of the adsorbents are shown in Fig. 3. Peaks of sodium at approximately 495 and 1069 eV, oxygen at approximately 528 and 976 eV, magnesium at approximately 303 eV, aluminum at approximately 71 and 116 eV, and silicon at approximately 99 and 150 eV were detected in RB because montmorillonite mainly consists of SiO₂, Al₂O₃, and MgO. The peak intensities of sodium significantly decreased following acid activation treatment, thereby indicating that the interlayer sodium ions were released from the RB due to the destruction of montmorillonite. Significant changes in other peaks were not observed after acid activation treatment. The concentration of the released sodium ions were approximately 2.2, 2.3, 2.2, 2.1, 2.2, 2.2, 1.8, 2.1, and 2.3 $\times 10^3$ mg/L in BS1, BS5, BS10, BN1, BN5, BN10, BP1, BP5, and BP10, respectively. These results are supported by the previously mentioned results.

Quality of Tap Water The quality of tap water was measured. The concentrations of sodium, potassium, magnesium, calcium, chloride, nitrate, and sulfate ions were 15.5, 2.4, 2.2, 12.3, 17.1, 4.3, and 12.2 mg/L, respectively. The electrical

conductivity and solution pH were 168.1 μ S/cm and 7.7, respectively. The results sufficiently satisfied the standard quality of tap water in Japan. The electrical conductivity in

Fig. 3. Binding Energies of Adsorbents

AXIS-NOVA instrument was used for measurement with an accelerating voltage 15 kV and electric current of 10 mA. (Color figure can be accessed in the online version.)

Table 2. Changes in Electrical Conductivity with the Adsorbents

Adsorbents	Electrical conductivity (μ S/cm)							
	Time (h)							
	0	0.17	0.5	1	2	6	22	24
RB	167.4	200.3	204.5	208.0	209.5	226.5	258.5	260.0
BS1	168.2	138.0	134.9	133.4	130.6	127.6	123.3	122.4
BS5	167.5	140.4	137.2	134.2	131.8	128.1	124.3	126.0
BS10	169.4	133.8	128.6	127.1	122.9	121.2	117.6	116.4
BN1	167.2	131.2	126.9	123.9	121.3	116.3	112.6	112.3
BN5	166.6	124.7	118.7	115.9	112.3	106.7	103.3	103.0
BN10	170.2	130.0	124.6	120.3	115.7	108.3	105.6	104.1
BP1	168.6	149.0	147.0	146.0	144.2	143.3	140.4	140.9
BP5	167.8	132.6	128.4	127.2	124.4	119.7	114.7	113.9
BP10	168.1	129.3	124.1	122.3	118.0	112.5	109.5	108.2

tap water was attributed to the previously mentioned ions and M-alkalinity, such as HCO_3^- and CO_3^{2-} .

Improvement in Tap Water Quality Using Acid Activated Bentonite First, we measured the changes in the electrical conductivity with adsorbents (Table 2). The values of the electrical conductivity in tap water before and after treatment with RB were 167.4 and 260.0 $\mu\text{S}/\text{cm}$, respectively, thereby indicating that sodium ions in the interlayer of montmorillonite were released from RB. Conversely, the values of the electrical conductivity in tap water ranged from 116.4–126.0, 103.0–112.3, and 108.2–140.9 $\mu\text{S}/\text{cm}$ with bentonite treated with sulfuric acid solution, nitric acid solution, and phos-

phoric acid solution, respectively. The results indicated that acid-activated bentonite is useful in decreasing the electrical conductivity of tap water. The relationship between the total removed ions and electrical conductivity is shown in Fig. 4. Based on the results, the correlation coefficient was -0.950 , thereby indicating that the removal of ions from the aqueous solution was useful in decreasing electrical conductivity.

Therefore, the changes in cation and anion concentrations were evaluated in detail. Specifically, BS10, BN10, and BP10 were selected for the experiment because the adsorbents exhibited a high potential for decreasing electrical conductivity in tap water. Figure 5 shows the changes in cation and anion concentrations. First, changes in the concentration of anions such as chloride ion, nitrate ion, and sulfate ion were not confirmed because each adsorbent did not exhibit a capability of ion exchange for anions in aqueous media. Conversely, RB, BS10, BN10, and BP10 exhibited the capability of removing cations. In the case of RB, significant numbers of magnesium ions, calcium ions, and potassium ions were removed within 10 min of commencing the experiment. However, the number of sodium ions released from RB simultaneously and significantly increased. RB mainly comprises montmorillonite, which belongs to the 2:1-type aluminosilicate. The basic structural unit of RB is composed of two tetrahedrally coordinated sheets of silicon ions surrounding sandwiched octahedrally coordinated sheets of aluminum ions. Additionally, RB has an adsorption capacity within its interlayer space and on the outer surface and edges.^{34,35} One of the adsorption mechanisms of cations such as magnesium ions, calcium ions, and potassium ions is ion exchange with sodium ions in the interlayer of the RB. Therefore, the concentration of sodium ions increased with increasing treatment time under our experimental condition. Finally, the electrical conductivity did not

Fig. 4. Relationship between Total Removed Ions and Electrical Conductivity

Amount adsorbent: 0.1 g, tap water volume: 100 mL, contact time: 0.17, 0.5, 1, 2, 6, 22, and 24 h, stirring: 250 rpm, temperature: room temperature. $\diamond$: RB, $\square$: BS, $\triangle$: BN, $\circ$: BP

Fig. 5. Changes in Cation and Anion Concentration

Amount adsorbent: 0.1 g, tap water volume: 100 mL, contact time: 0.17, 0.5, 1, 2, 6, 22, and 24 h, stirring: 250 rpm, temperature: room temperature. $\diamond$: RB, $\square$: BS10, $\triangle$: BN10, $\bullet$: BP10 (Color figure can be accessed in the online version.)

Table 3. Changes in pH with the Adsorbents

Adsorbents	pH							
	Time (h)							
	0	0.17	0.5	1	2	6	22	24
RB	7.5	8.0	8.1	8.1	8.1	8.1	7.9	7.9
BS1	7.7	7.4	7.3	7.3	7.3	7.3	7.3	7.3
BS5	8.1	8.0	7.9	7.9	7.9	7.8	7.7	7.8
BS10	7.9	7.7	7.7	7.6	7.5	7.6	7.5	7.3
BN1	7.7	7.5	7.3	7.4	7.4	7.3	7.2	7.2
BN5	7.6	7.2	7.2	7.1	7.1	7.0	6.9	6.8
BN10	7.5	7.5	7.4	7.3	7.3	7.3	7.3	7.2
BP1	7.4	7.5	7.4	7.5	7.4	7.4	7.4	7.5
BP5	7.5	7.4	7.4	7.3	7.3	7.3	7.3	7.2
BP10	7.8	7.5	7.3	7.3	7.3	7.2	7.2	7.0

decrease with the use of RB. Additionally, BS10, BN10, and BP10 exhibited an efficient capability for removing cations. Specifically, the number of magnesium ions, calcium ions, and potassium ions that were removed increased with an increase in the treatment time. Additionally, the number of sodium ions that were released was not confirmed using the adsorbents, thereby indicating that the electrical conductivity decreased. Therefore, acid activation of bentonite using sulfuric acid, nitric acid, and phosphoric acid solution constitutes a considerably useful technique for decreasing electrical conductivity in tap water. In comparison of the decrease in electrical conductivity among BS10, BN10, and BP10, the removal percentage was in the order $BS10 < BP10 < BN10$ under our experimental condition. The values of pore volume ($2 < d \leq 50$ nm) and CEC in BN10 were larger than those in BP10 and BS10. Thus, the results indicate that pore volume and CEC are useful factors in decreasing electrical conductivity when compared to the specific surface area among BS10, BN10, and BP10. However, further studies are required to elucidate the physicochemical factors of the adsorbent to control the decrease in the electrical conductivity of tap water. The pH of the solution was measured before and after treatment (Table 3). Significant changes in pH levels were not observed with adsorbents (between approximately 7.0 and 8.0). Therefore, the application of adsorbents to decrease the electrical conductivity of tap water during wire electrical discharge machining can be useful in controlling the electrical conductivity of tap water.

Finally, various previous studies have investigated the adsorption method of heavy metal ions such as lead ion and copper ions and organic polymer compounds such as humic acid and dye using acid-activated bentonite.^{15,20,36–38} These studies have reported that acid-activated bentonite is an useful adsorbent for removal of toxic compounds in aqueous media. Subsequently, these techniques contributed toward the purification of wastewater. Therefore, there is a possibility that the developed acid-activated bentonite in this study contributes to wastewater purification. However, further studies are necessary to elucidate the adsorption capability of heavy metals and/or organic polymer compounds in detail from aqueous media.

Conclusion

In the study, acid-activated bentonite using sulfuric acid, nitric acid, and phosphoric acid solution was prepared, denoted

as BS10, BN10, and BP10, respectively, and the adsorption capability of ions from tap water was assessed. The specific surface area and mesopore volume ($2 < d \leq 50$ nm) of BS10, BN10, and BP10 exceeded those of RB. The results indicated that the acid activation of bentonite with an acidic solution increases the specific surface area and pore volume. Additionally, the changes in the electrical conductivity and cation/anion concentration were evaluated. The electrical conductivity of tap water decreased by approximately 31, 39, and 36% due to BS10, BN10, and BP10, respectively. The relationship between the concentrations of the removed ions and electrical conductivity was presented, and the correlation coefficient corresponded to -0.950 . Furthermore, a significant number of magnesium ions, calcium ions, potassium ions, and sodium ions were removed after treatment using acid-activated bentonite when compared to that using RB. Therefore, BS10, BN10, and BP10 were useful in controlling the reduction in the electrical conductivity of tap water.

Conflict of Interest The authors declare no conflict of interest.

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