Sustainable utilisation of water treatment residue as a porous geopolymer for iron removal from groundwater

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Abstract

Raw water is significant resources for industrial water usage, but this water is not directly suitable for use due to the presence of contaminants. Therefore, pre-treatment is essential. In addition, the presence of iron (Fe) and manganese (Mn) in groundwater can result in a reddish-brown colour and undesirable taste and odour. The treatment generates water treatment residue (WTR) which consists of silt, clay and undesirable components. Most WTR is conventionally disposed of in landfill. A number of expensive and complex technologies are being used for the removal of such iron and manganese. Due to the high Al_2O_3 and SiO_2 content in WTR, the use of WTR-based geopolymers for Fe/Mn removal is proposed in this study. With the availability of free alkali in the geopolymer framework, the OH-releasing behaviour of the WTR-based geopolymer was investigated by the precipitation of Fe(II) ion. The WTR-based geopolymer was calcined at 400°C and 600°C to obtain a strong geopolymer matrix with the ability to remove Fe/Mn ions. The results show that the WTR-based geopolymer has the potential to remove Fe from Fe-contaminated water. Hydroxide ions are released from the geopolymer and form an $\text{Fe}(\text{OH})_3$ precipitate. A calcination temperature of 400°C provides total removal of the Fe after 24 h of immersion. In addition, the existence of $\text{Fe}(\text{OH})_3$ helps to coprecipitate the $\text{Mn}(\text{OH})_2$ in the Fe/Mn solution leading to a significant reduction of Mn from the solution. The pH value and retention time play an important role in the final metal concentration. The final pH of the solution is close to 8.5, which is the recommended value for boiler water. This method offers an alternative use of WTR in making a porous geopolymer for groundwater Fe removal using a simple method.

1. Introduction

Raw water is the main resource for industrial water usage for washing, fabricating, processing, diluting, boiling and cooling products. Raw water includes rainwater, groundwater, and water from rivers and lakes. This water is not directly used due to the presence of contaminants, and pre-treatment of raw water is essential [1]. Aluminium sulphate (Alum) and other coagulants, such as polymers, are generally used to coagulate the fine suspended solids in raw water by forming a mass large enough to settle and enable removal [2]. This water treatment residue (WTR) consists of silt, sand, clay, organic substances, and undesirable components with a very high water content of up to 80% [3]. In Thailand, 137,900 tonnes of WTR is generated annually and is conventionally disposed of in landfill [4]. Attempts have been made to use WTR as a replacement for fire brick clay since its properties are similar to clay. The addition of 10% WTR with a firing temperature above 1000°C has been shown to obtain good properties of water absorption and mechanical resistance [5]. However, using a large amount of WTR as a clay replacement in ceramic production is problematic due to its seasonally dependent availability [4, 5].

Groundwater is a major resource for tap and drinking water. However, it contains iron and manganese contaminants due to leaching from the laterite soil layer after heavy rainfall with alternating wet and dry periods [6]. In particular, the presence of iron can contaminate water, resulting in a reddish-brown colour and undesirable taste and odour. The dissolved, colourless form of iron, Fe(II), is converted into an

insoluble form, Fe(III), in contact with air. In groundwater, iron exists in Fe(II) form due to the lack of oxygen. When exposed to air in a pressure tank or the atmosphere, a cloudy and reddish-brown substance is formed [5, 6]. In addition, dissolved manganese may form black particulates in water. These substances can deposit in the water pipe and subsequently flow through the tap, leading to reddish-brown stains on tableware and laundry that are difficult to remove. Iron and manganese removal systems are therefore required to overcome this problem. Many advanced materials are available to remove iron from water; for example, nano bentonite [7], microporous chitosan blend membrane [8], and carbon nanotubes [9]; however, some technologies are expensive and not cost-effective for water treatment systems.

Several studies report the use of waste ash in a geopolymer, reflecting its suitability and availability [10, 11]. Alkaline solutions and heat curing are used to activate the reaction between the active alumina (Al_2O_3) and silica (SiO_2) in the ash, leading to the formation of Si-O-Al bonds in an alumino-silicate product. Geopolymeric materials are the alternative choice for recycling industrial waste [12] and are often used as materials for roadway construction and pavements due to their high disposal rate [13, 14, 15]. Apart from waste ash, natural clay and calcined clay have also been widely used as source materials for geopolymer synthesis [16, 17, 18]. Clay is rich in Al_2O_3 and SiO_2 , and is classified as an alumino-silicate material and is normally calcined to improve its reactivity. Generally, geopolymer materials are used as construction materials; however, since WTR has a high content of Al_2O_3 and SiO_2 and contains clay, porous geopolymers have recently been proposed as a CO_2 absorbent [19], acidic-wastewater treatment [20], and heavy metal remover [21, 22]. Therefore, this research proposes the use of WTR as the source material for geopolymer production. Given the free alkali content of the geopolymer framework, the OH-releasing behaviour of a WTR-based geopolymer was investigated based on the precipitation of Fe(II) ions. The characteristics of Fe(II) removal were studied through heating a WTR-based geopolymer. This alternative use of WTR in forming a geopolymer, and its use for iron removal from groundwater, is simple and cost-effective.

2. Materials And Methods

2.1 Materials

The WTR came from the raw water treatment system of a power plant in Rayong, Thailand. This residue has high moisture content and was therefore oven-dried at 115°C overnight and then ground and sieved through sieve no. 100 (146-micron opening) for use as the raw material in geopolymer production. The grinding process reduces the grain size and promotes amorphisation, resulting in the enhancement of the reactivity of the materials [11]. The WTR powder was used without calcination since an alkali-releasing application is required rather than a high-strength building application. 10 M sodium hydroxide solution (10 M NaOH) and sodium silicate (Na_2SiO_3) were used to activate the geopolymerisation. The Na_2SiO_3 solution was composed of soluble SiO_2 (30 wt%), Na_2O (9 wt%), and H_2O (61 wt%). Aluminium powder was used as a pore generator and propylene glycol as a pore refiner [19].

Iron (II) sulphate heptahydrate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$, MW = 278.03 g mol⁻¹) was used to prepare the 100-ppm stock solution. Deionized (DI) water was used throughout the experiment. The pH of the Fe solution was 4.5. In addition, manganese (II) nitrate tetrahydrate ($\text{Mn}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$, MW = 251.01 g mol⁻¹) was also dissolved in DI water to obtain a 100-ppm Mn solution for the study of the common ion effect. The pH of the Mn solution was 4.9.

2.2 Porous geopolymer preparation

The mix proportions of porous geopolymer (PG) are shown in Table 1. The ground WTR powder was thoroughly mixed with Al powder (0.25% by weight of WTR). Na_2SiO_3 solution and 10 M NaOH were homogeneously mixed and added to the blended powder to obtain a uniform paste, followed by the PG (0.25% by weight of WTR). An $\text{Na}_2\text{O} \cdot \text{SiO}_2$ -to-NaOH ratio of 9 : 1 was selected to increase the free alkali content. The paste was poured into a doughnut-shaped silicon mould (0.5-cm ID, 2-cm OD) and oven-cured at 115°C for three hours. After that, the effect of the heat treatment on the geopolymer’s stability and reactivity was studied by calcination of the geopolymer at 400°C and 600°C for three hours. The samples were identified as Geo115 (no calcination), Geo400 (400°C calcination), and Geo600 (600°C calcination). Water absorption of the geopolymers was measured by immersion of a weighted sample in DI water for 24 h. An increase in weight was used to calculate the percentage of water absorption. The pH of the immersed water was also measured. In addition, small samples were used for porosity measurement with a mercury intrusion porosimeter (MIP, Micromeritics AutoPore V).

Table 1 Mix proportion of the geopolymer					
Samples	WTR (g)	Na_2SiO_3 (g)	10 M NaOH (g)	Al (g)	PG (g)
WTR-geopolymer	50	45	5	0.25	0.25

2.3 Fe removal

Three geopolymers (i.e., Geo115, Geo400, Geo600) were examined for Fe removal from the aqueous solution with a metal concentration of 100 ppm (mg L⁻¹). The metal ion concentration in wastewater was found to be 1–100 ppm [23]. The geopolymer-to-Fe solution ratio was 1 : 100 w/v (g mL⁻¹) [22]. The geopolymer and Fe solution were put in a beaker and stirred continuously with a magnetic stirrer, and the pH was monitored. The beaker was covered with a watch glass to avoid water evaporation during the experiment. The Fe concentration in the solution was monitored using a selective colourimeter with specific reagents (Hanna HI721). The reagents were used to form colour complexes according to the EPA phenanthroline method 315B. The reaction between iron (Fe(II)) and the 1,10-phenanthroline caused an orange tint. After the metal removal, the amounts of silica and aluminium leachates were also measured using selective silica and aluminium colourimeters (Hanna HI705 and HI96712). The silica reagent was tested in accordance with ASTM D859 (heteropoly blue method), and the aluminium reagent was tested in accordance with the aluminon method (reddish tint) [24].

2.4 Fe/Mn removal

The Geo400 sample was selected to study the effect of coprecipitation on Fe(II) removal. Since manganese (Mn) generally co-occurs in raw water, the Mn(II) form was selected as the common ion. The Mn concentration in the solution was measured using a selective colourimeter with specific reagents (Hanna HI709). The reagents were used to form colour complexes according to the standard methods for the examination of water and wastewater (periodate method). The reaction between the reagents and manganese caused a pink tint in the sample.

In addition, a mixture of a 100-ppm Fe/Mn solution was studied using a geopolymer-to-solution ratio of 1:50 and 1:100 w/v. The pH and concentration of Fe and Mn were investigated after a retention time of 24 h (and 48 h, if applicable). Concentrations of SiO₂ and Al were also measured.

3. Results And Discussion

3.1 Characteristics of WTR and geopolymer

Chemical compositions of WTR as shown in Table 2 were measured in terms of oxide forms using an X-ray fluorescence spectrometer (XRF, Rigaku ZSX Primus). The main constituents were 46.2% SiO₂ and 40.0% Al₂O₃. SiO₂ and Al₂O₃ were from minerals in soil, and additional Al₂O₃ was from the alum used in coagulation of the fine residual [24]. Large amounts of SiO₂ and Al₂O₃ imply that this residual is possibly a good source material for geopolymer production.

Table 2
Chemical compositions of WTR

Composition (%)	SiO ₂	Al ₂ O ₃	CaO	Fe ₂ O ₃	SO ₃	MgO	K ₂ O	MnO	others
WTR	46.2	40.0	1.8	4.3	1.0	0.6	1.6	1.4	3.1

Figure 1 shows the doughnut-shaped geopolymer (Geo115 sample) before calcination at 400°C and 600°C. After calcination, samples of a darker colour were obtained. When the geopolymers were immersed in DI water for 24 h, the colour of the water changed depending on the calcined temperature of the geopolymer. Geo115 showed the darkest colour, followed in turn by Geo400 and Geo600. With a high calcination temperature of 600°C, a strong geopolymer matrix was achieved resulting in water of a clear colour. The pH of the immersed water was 11.0, 9.6 and 9.6 for Geo115, Geo400 and Geo600, respectively, as shown in Table 3. This result implies that the dissolved compounds and OH-ion were easily leached out from the geopolymer with a curing temperature of 115°C and a short heating time of three hours. It has been reported that the use of boric acid can overcome this problem, since boric acid enables the formation of oxygen bridges in the silicate structure, resulting in greater stability [20]. The results of pore analysis of the geopolymers using a MIP are shown in Table 3. It can be seen that the total pore volume increased with calcination temperatures from 0.38, 0.47, to 0.58 for Geo115, Geo400 and Geo600, respectively. The increase in porosity with increased temperature was consistent with the increase in water absorption of the geopolymer. The calcination process resulted in increased numbers of

pores due to the expansion of sodium silicate in the geopolymer mixture [25]. The bulk density was reduced from 1.20 to 0.97 g mL⁻¹ with the increase in calcination temperature. In addition, the higher total pore area was obtained with the increase in the calcined temperature; the Geo400 pore area was highest at 24.69 m² g⁻¹. Therefore, the calcination process provided higher porosity leading to higher water absorption than that of the non-calcined sample (Geo115).

Table 3
MIP result and water absorption of geopolymer

Properties	Geo115	Geo400	Geo600
Total volume (mL g ⁻¹)	0.34	0.47	0.58
Total pore area (m ² g ⁻¹)	0.09	24.69	21.37
Bulk density (g mL ⁻¹)	1.20	1.04	0.97
Porosity (%)	40.73	49.43	56.51
Water absorption (%)	20.30	29.58	31.73
pH of DI water (after 24-h immersion)	11.0	9.6	9.6

The morphology of the WTR and geopolymers was studied by X-ray powder diffractometer (XRD, Panalytical/Expert) and the results are shown in Fig. 2. The XRF detected peaks of SiO₂, Al₂O₃, and CaO in the WTR. The XRD patterns of Geo115 were slightly different, showing a lower peak height of SiO₂ at 21 and 37 °2theta compared to that of the WTR. Peak shift was not significantly detected due to the low reactivity of the WTR. The calcination of WTR is therefore suggested to improve its reactivity [12]. A peak of kaolinite was identified at 20 °2theta owing to the presence of an alumino-silicate compound in the residual.

For the Geo400 and Geo600 samples, calcination of the geopolymer at 400°C and 600°C reduced the SiO₂ peak at 12 °2theta, kaolinite at 20 °2theta, and Al₂O₃ at 25 °2theta. For calcination at 600°C, the Geo600 sample had a significant peak of albite at 20 °2theta, showing the crystallinity of the material [11]. Albite is a zeolite formed by the reaction of the alumino-silicate compound in WTR and alkali solutions under heat curing. Strong bonding in the geopolymer is achieved in the same way as for a ceramic material. Some peaks were, however, still located at the same positions as those of WTR, indicating unreacted compounds. Peak areas in the geopolymer were calculated between 18 and 34 °2theta using the Origin program, and the results are shown in Table 4. Calcination increased the area of the peak, implying a higher degree of reaction in the geopolymers.

Table 4
Peak areas of XRD patterns

Samples	2theta (Degree)	Area (a.u.)
WTR (Raw materials)	16.06–34.04	6654.3
Geo115	16.01–34.02	7105.9
Geo400	16.01–34.02	7306.4
Geo600	15.94–34.04	7933.0

The microstructures of the geopolymers obtained from scanning electron microscopy (SEM, Leo 1450VP, gold coating) are shown in Fig. 3. The flaky shape of the WTR was still observed in the Geo115 sample. When the geopolymer was calcined at 400°C and 600°C, a dense and strong matrix was observed in the geopolymer samples. In addition, more pores were generated due to the expansion of sodium silicate at high temperatures [25]. Thus, increases in pore volume and surface area were achieved.

3.2 Fe removal by geopolymers

3.2.1 Fe removal system

Figure 4 shows the Fe concentration plotted against the retention time of the Fe-contaminated solutions containing each of the three geopolymers. Geo115 had a lower Fe reduction capability than the others. Considering the final pH (Table 5), the Geo115 sample had a high pH of 9.6, at which the iron hydroxide was redissolved into the solution. The Geo400 sample removed all the Fe in the aqueous solution in 24 h, whereas Geo600 achieved approximately 50% removal (45.0 ppm remaining). Geo600 had a dense matrix; therefore, its OH-releasing ability was less than that of Geo400. The final pH of the solution containing Geo600 was only 6.5. Generally, Fe(II) converts to Fe(III) and precipitates at around pH 5. The plot of pH against percentage Fe removal for the Geo400 removal system is shown in Fig. 5. Total Fe removal was achieved when the pH value reached 8. During the precipitation process, $\text{Fe}(\text{OH})_3$ was formed as an orange substance suspended in the aqueous solution. Some of the substance was also deposited on the geopolymer surface, which was the release site of the hydroxide ions (OH ions) originating from the alkali solution in the geopolymer mixture. This substance can be totally removed from the solution by filtration, as shown in Fig. 6. Only the colour of the Geo115-solution was not removed by filtration, since it was the colour of the mineral leached from the WTR. In addition, Si and Al ions were detected in the leaching solution. With Geo115, high amounts of Si and Al were leached out; these metals were reduced by calcination owing to the strong geopolymer matrix.

Table 5
Conditions and ion concentrations in the Fe-aqueous solution

Ion removal	Samples	Solid:solution ratio	Retention time (h)	Initial pH	Final pH	Final concentrations (ppm)			
						Fe	Mn	Si	Al
Fe	Geo115	1 : 100	48	4.5	9.6	45.0	-	130	10
	Geo400	1 : 100	24	4.6	8.3	0	-	15	20
	Geo600	1 : 100	48	4.4	6.5	49.0	-	15	15

The functional group of the orange substance deposited on the geopolymer was characterised using attenuated total reflectance-Fourier transform infrared spectroscopy (ATR-FTIR, Perkin Elmer Frontier). The FTIR spectra are shown in Fig. 7. It was found that the substance deposited from Geo115 had a dominant peak of Fe-O stretching vibrational mode at 580 cm^{-1} and of Fe-O bending vibrational mode at 1020 cm^{-1} [26]. An Fe-O-Si peak was also noticeable at 1050 cm^{-1} . Since there were high amounts of Fe (Fe^{2+}) and SiO_2 in the immersed solution of the Geo115 system (Table 5), $\text{Fe}(\text{OH})_3$ can react with Fe^{2+} ion leading to the formation of magnetite (Fe_3O_4) [27]. These Fe_3O_4 particles are formed and covered with the SiO_2 layer resulting in the strong vibration of the Fe-O-Si bond [28].

3.2.2 Combined Fe(II) and Mn(II) removal

Mn removal system

Manganese (Mn) is also found in raw water; Geo400 was selected to remove the Mn owing to its efficiency in removing Fe. The solid-to-solution ratio of 1 : 100 and a 100-ppm solution of Mn was used according to a previous section. The result as reported in Table 6 show that the solid-to-solution ratio of 1 : 100 did not achieve significant Mn removal, with 94.0 ppm remaining at 24 h. Little OH was released from the Geo400, and the solubility product constant (K_{sp}) of $\text{Mn}(\text{OH})_2$ was 2×10^{-13} higher than that for $\text{Fe}(\text{OH})_3$. The K_{sp} of $\text{Fe}(\text{OH})_3$ was 4×10^{-38} ; therefore, $\text{Fe}(\text{OH})_3$ was easier to precipitate than $\text{Mn}(\text{OH})_2$.

Table 6
Conditions and ion concentrations in the Mn- and Fe/Mn-aqueous solutions

Ion removal	Samples	Solid:solution ratio	Retention time (h)	Initial pH	Final pH	Final concentrations (ppm)			
						Fe	Mn	Si	Al
Mn	Geo400	1 : 100	24	4.9	6.8	-	94.0	20	15
		1 : 50	24	4.9	7.7	-	14.0	45	25
Fe/Mn	Geo400	1 : 100	24	4.9	6.0	11.8	40.0	10	20
		1 : 50	24	4.9	8.1	0	2.0	0	15

The solid-to-solution ratio was then changed to 1 : 50. The final pH of the solution was increased from 6.8 (at a ratio of 1 : 100) to 7.7, leading to increased $\text{Mn}(\text{OH})_2$ precipitation. The Mn concentration was reduced to 14.0 ppm. More Si and Al were leached out to the solution due to its increased alkalinity. Generally, Mn ions are treated by adjusting the pH to 10; however, this process consumes a substantial amount of alkali [29]. Therefore, the use of a geopolymer can reduce the required pH, providing Mn removal and efficient WTR utilisation.

Fe/Mn removal system

In this system, a mixed solution of Fe and Mn was used to study the effect of the coprecipitation of Fe and Mn hydroxide. The results reveal that Mn precipitation was greater in the mixed Fe/Mn solution. At a solid-to-solution ratio of 1:100, 11.8 ppm of Fe and 40.0 ppm of Mn remained. At a solid-to-solution ratio of 1:50 (Table 6), Fe was totally removed and 2.0 ppm Mn remained. The pH of the solution increased from 6.0 to 8.1. It has been reported that Mn(II) can be removed from an aqueous solution by adsorption and coprecipitation with Fe ions [29]. The addition of alkali (OH^- ions from Geo400) increased the pH of the solution, leading to an increase in Fe precipitate and the Mn removal ratio. Van der Waals force between the $\text{Mn}(\text{OH})_2$ and $\text{Fe}(\text{OH})_3$ molecules also played an important role in agglomeration [22]. Researchers have shown that the precipitation of iron hydroxide ($\text{Fe}(\text{OH})_3$) can be used to remove heavy metals (e.g., arsenic, lead, nickel) in wastewater, as $\text{Fe}(\text{OH})_3$ absorbs these heavy metals onto its surface [30]. These metals can exist in both neutral and anionic forms depending on the pH; hence, precipitation of the metal is pH dependent. In addition, Si was completely removed, conforming to the FTIR results showing that Si formed bonds with $\text{Fe}(\text{OH})_3$. In addition, some Si was absorbed onto the $\text{Fe}(\text{OH})_3$ surface leading to a reduced Si content of the solution. Fe and Mn values after treatment with Geo400 are suitable for use in industrial applications, e.g., material and machine washing, or water supply for boilers.

3.3 Comparison of the Fe removal materials

From the results of this study, it can be seen that geopolymers have the potential to be utilised to remove Fe from contaminated water, an environmentally positive use of WTR. However, as presented in Table 7, the comparison between this geopolymer (Geo400) and various materials on Fe removal indicates that the porous geopolymer-precipitation process has high efficiency of Fe removal but the operation time is slightly longer than the adsorption process.

Table 7
Comparison of Fe removal materials between geopolymer and various materials

Materials	Raw materials	Fe removal process	Conditions for Fe removal	Efficiency	References
Geo400	WTR	Precipitation	1:100 w/v 100-ppm Fe 24 h	100 %	This work
Fe(OH) ₃ flocs	FeCl ₃ + ammonia solution	Adsorption	1:100 w/v 5-ppm Fe 100 min	98 %	Ref. [6]
A.ferrooxidans	A.ferrooxidans (Bacteria)	Coprecipitation	1:100 v/v 9,000-ppm Fe 100 h	33 %	Ref. [29]
Activated carbon	Coconut shells	Adsorption	1:50 w/v 9,000-ppm Fe 120 min	55 %	Ref. [31]

This geopolymer could be applied for Fe removal in groundwater treatment systems. The retention time is essential for the precipitation process from OH release. The pH value of the solution is increased with precipitation of the metal hydroxide. A coagulant is recommended in some cases; however, the cost of the chemical should be considered. Fe(OH)₃ precipitate can be filtered off by sand filtering. The final pH of the solution is in the range 5.5–9.0, which complies with a universal standard pH for industrial discharge water. In addition, a common recommendation is to maintain the pH value of boiler water at 8.5, since acidic water is corrosive and alkaline water is prone to scaling.

4. Conclusions

A WTR-based geopolymer has the potential to remove Fe in Fe-contaminated water using the precipitation method. Calcination at 400°C creates a strong matrix in the composite. Hydroxide ion from the geopolymer is released and forms Fe(OH)₃ precipitate. The reaction between Fe(OH)₃ and Fe²⁺ ion forms Fe₃O₄, which was subsequently covered with the SiO₂ layer resulting in the Fe-O-Si bond. Geo440 completely removed the Fe after 24-h immersion. In addition, the existence of Fe(OH)₃ helps coprecipitate the Mn(OH)₂ in an Fe/Mn solution leading to a significant reduction in Mn from the solution. The pH

value and retention time play an important role in the final metal concentration. The final pH of the solution was close to 8.5, which is the recommended value for boiler water. Fe removal with a WTR-based geopolymer is suitable for groundwater treatment. Industrial waste can be beneficially utilised leading to an environmentally friendly water treatment solution.

Declarations

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Authors' contributions

Rattanasasak, U. designed, directed and coordinated this research. Pachana, KP., Nuithitikul, K., Jitsangiam, P., and Chindaprasirt, P. provided conceptual and technical guidance for all aspects of the project. All authors reviewed, edited, read and approved the final manuscript.

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Availability of data and materials

All data generated or analysed during this study are included in this published article.

Competing interests

The authors declare they have no competing interests.

References

1. Clever M, Jordt F, Knauf R, Rübiger N, Rüdebusch M, Hilker-Scheibel R. Process water production from river water by ultrafiltration and reverse osmosis. *Desalin* 2000;131(1–3):325–336.
2. Matilainen A, Vepsäläinen M, Sillanpää M. Natural organic matter removal by coagulation during drinking water treatment: a review. *Adv Colloid Interface Sci* 2010;159(2):189–197.
3. Tantawy MA. Characterization and pozzolanic properties of calcined alum sludge. *Mater Res Bull* 2015;61:415–421.
4. Prakongwittaya W, Asavapisit S, Piyapanuwat R, Muhummud T. Effect of seasons and sources of raw water on the properties of water treatment residue compacted clay. *Int J. Environ Sci Dev*

- 2017;8(5):337–341.
5. Sarabia A, Sanchez J, Sanchez JV. Effect of the incorporation of residual sludge from water treatment on the technological properties of ceramic bodies: A review. *J. Phys Conf Ser* 2019;1388:012018.
 6. Abraham N, James J, Banerji T, Menon R. Development of a novel groundwater iron removal system using adsorptive Fe(II) process. *Groundwater Sustainable Dev* 2020;10:100318.
 7. Mohamed AM, Ayad MI, Eledkawy MA, Darweesh MA, Elmelegy EA. Removal of iron, zinc, and nickel ions using nano bentonite and its applications on power station wastewater. *Heliyon* 2021;7(2):e06315.
 8. Reiad NA, Salam OM, Abadir E, Harraz F. Adsorptive removal of iron and manganese ions from aqueous solutions with microporous chitosan/polyethylene glycol blend membrane. *J. Environ Sci* 2012;24(8):1425–1432.
 9. Alimohammadi V, Sedighi M, Jabbari E. Experimental study on efficient removal of total iron from wastewater using magnetic-modified multi-walled carbon nanotubes. *Ecol Eng* 2017;102:90–97.
 10. Chindaprasirt P, Jenjirapanya S, Rattanasak U. Characterizations of FBC/PCC fly ash geopolymeric composites. *Constr Build Mater* 2014;66(1):72–78.
 11. Eiamwijit M, Pachana K, Kaewpirom S, Rattanasak U, Chindaprasirt P. Comparative study on morphology of ground sub-bituminous FBC fly ash geopolymeric material. *Adv Powder Technol* 2015;26(4):1053–1057.
 12. Chindaprasirt P, Rattanasak U, Vongvoradit P, Jenjirapanya S. Thermal treatment and utilization of Al-rich waste in high calcium fly ash geopolymeric materials. *Int J. Miner Metall Mater* 2012;19(9):872–878.
 13. Jitsanigam P, Biswas WK, Compton M. Sustainable utilization of lime kiln dust as active filler in hot mix asphalt with moisture damage resistance. *Sustainable Mater Technol* 2018;17:e00071.
 14. Nusit K, Jitsangiam P, Kodikara J, Bui HH, Leung GLM. Advanced characteristics of cement-treated materials with respect to strength performance and damage evolution. *J. Mater Civ Eng* 2017;29(4):04016255.
 15. Purwana YM, Nikraz H, Jitsangiam P. Experimental study of suction-monitored CBR test on sand-kaolin clay mixture. *Int J. GEOMATE* 2012;32(2):419–422.
 16. Cheng T.W, Lee M.L, Ko M.S, Ueng T.H, Yang S.F. The heavy metal adsorption characteristics on metakaolin-based geopolymer. *Appl Clay Sci* 2012;56: 90–96.
 17. Amin N, Faisal M, Muhammad K, Gul S. Synthesis and characterization of geopolymer from bagasse bottom ash, waste of sugar industries and naturally available china clay. *J. Cleaner Prod* 2016;129:491–495.
 18. Tippayasam C, Kaewpapasson P, Thavorniti P, Panyathanmaporn T, Leonelli C, Chaysuwan D. Effect of Thai kaolin on properties of agricultural ash blended geopolymers. *Constr Build Mater* 2014;51:455–459.

19. Chindaprasirt P, Rattanasak U. Characterization of the porous alkali-activated fly ash composite as solid adsorbent. *Int J. Greenhouse Gas Control* 2019;85:30–35.
20. Chindaprasirt P, Rattanasak U. Synthesis of porous alkali-activated materials for high-acidic wastewater treatment. *J. Water Process Eng* 2020;33:101118.
21. Muzek MN, Svilovic S, Ugrina M, Zelic J. Removal of copper and cobalt ions by fly ash-based geopolymer from solution-equilibrium study. *Desalin. Water Treat* 2016;57(23):10689–10699.
22. Pachana PK, Rattanasak U, Jitsangiam P, Chindaprasirt P. Alkali-activated material synthesized from palm oil fuel ash for Cu/Zn ion removal from aqueous solutions. *J. Mater Res Technol* 2021;13:440–448.
23. Pugazhenthiran N, Anandan S, Ashokkumar M. Removal of heavy metal from wastewater. In *Handbook of Ultrasonics and Sonochemistry*. Singapore: Springer; 2016. DOI: 10.1007/978-981-287-278-4_58
24. APHA. Standard methods for the examination of water and wastewater. 18th ed. American Public Health Association, 1992.
25. Haq EU, Padmanabhan SK, Licciulli A. Microwave synthesis of thermal insulation foams from coal derived bottom ash. *Fuel Process Technol* 2015;130:263–267.
26. Zhang T, Zhao N, Li J, Gong H, An T, Zhao F, Ma H. Thermal behavior of nitrocellulose-based superthermites: effects of nano-Fe₂O₃ with three morphologies. *RSC Adv* 2017;7:23583–23590.
27. Lenders JJ, Mirabello G, Sommerdijk NAJM. Bioinspired magnetite synthesis via solid precursor phases. *Chem Sci* 2013;7:5624.
28. Quy DV, Hieu NM, Tra PT, Nam NH, Hai NH, Thai Son N, Nghia PT, Anh NTV, Hong TT, Luong NH. Synthesis of silica-coated magnetic nanoparticles and application in the detection of pathogenic viruses. *J. Nanomater* 2013;2013:603940.
29. Yoo K, Sasaki K, Hiroyoshi N, Tsunekawa M. Removal of Mn Ions by Biological Coprecipitation of Fe Ions. *Geosyst Eng* 2010;13(3):91–96.
30. Wang JW, Bejan D, Bunce NJ. Removal of arsenic from synthetic acid mine drainage by electrochemical pH adjustment and coprecipitation with iron hydroxide. *Environ Sci Technol* 2003;37:4500–4506.
31. Nandeshwar SN, Mahakalakar AS, Gupta RR, Kyzas GZ. Green activated carbons from different waste materials for the removal of iron from real wastewater samples of Nag River. *India J. Mol Liq* 2016;216:688–692.

Figures

Figure 1

Geopolymers: (a) doughnut-shape Geo115; (b) colour of water after 24-h immersion

Figure 2

XRD patterns of WTR and geopolymers

Figure 3

Figure 4

Fe concentration of the solutions and the retention time

Figure 5

pH profile of the Fe-contaminated solution with Geo400 with a 24-h retention time

Figure 6

Colour of Fe-contaminated solution after the removal process

Figure 7

FTIR spectra of deposited substance covering the geopolymer surface