Novel Multifunctional Sewage Sludge-based Adsorbents For Treatment Of Municipal Wastewater

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Common adsorbents are typically developed to treat a sole targeted pollutant in wastewater. This research provides the multifunctional features of ceramsite, a sewage sludge-based adsorbents developed for removing phosphorus (P) and disinfecting pathogens in municipal wastewater. The ceramsite adsorbents were made by mixing sewage sludge with zeolite, bentonite, and magnesium oxide (MgO) powders. Batch experiment results revealed that the developed ceramsite adsorbents significantly removed phosphorus (0.45mgP/g), effectively inactivated fecal coliforms (3.7 log reduction). and reached an optimum hydraulic retention time (HRT) of 6 hours. Moreover, in the continuous filter columns, the developed adsorbents effectively removed P(0.14mgP/g : 93%) and simultaneously inactivated fecal coliforms (3.4 log reduction) in wastewater treatment plant effluent. The ratio of added MgO powder and pH values correlated well with phosphorus and pathogen removal results $(R^2 > 0.96)$. The adsorption isotherm for TP removal by the adsorbents (S50 and S60) fitted well with the Langmuir model $(R^2 > 0.95)$. The kinetics of TP and pathogen removal by the adsorbents followed the pseudo-second order $(R^2 > 0.97)$ the first-order reaction models $(R^2 > 0.87)$, respectively.

Keywords: Adsorbents; Sewage sludge; Municipal wastewater; Phosphorus; Pathogens
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1. Introduction

Municipal wastewater containing high concentrations of organics, nutrients and pathogens is conventionally treated by biological nutrient removal systems, such as step feed activated sludge, oxidation ditch (OD), modified ludzack ettinger (MLE) with an upfront anaerobic reactor, and sequencing batch reactor (SBR). These treatment systems typically employ biological processes e.g., biological oxidation, nitrification and denitrification for removing organics and nitrogen (N), respectively in the wastewater. However, these processes were not effective in phosphorus (P) removal and pathogenic inactivation. In particular, because of phosphorus shortage crisis [1] and high phosphorus concentrations in the aquatic environments causing eutrophication problems, removal and recovery of P from municipal

wastewater have currently become a significant issue [2, 3]. Besides, due to ineffective treatment performance for inactivating pathogens by the conventional wastewater treatment systems, pathogens in the treated wastewater have been found to cause many health risks for the people [4].

Various biological treatment processes, such as constructed wetlands, anaerobic, anoxic, and aerobic reactors [5], algal-bacterial assimilation [6], and enhanced biological phosphorus removal (EBPR) [7], have been employed to remove pollutants, especially phosphorus (P) from wastewater. However, those treatment systems could be generally energy-intensive, costly, requiring larger footprint, and often generating excess sludge and residue biomass that require further disposal [3, 8, 9]. Although some of the generated sludge from wastewater treatment plants has been directly reused as fertilizers, more proper and sustainable management in an environmentally responsible way is still required [10]. Additionally, precipitation [11], ion exchange [12], membrane filtration [13], electrodialysis [14], and magnetic separation [15] have also been utilized as physico-chemical techniques for P removal from wastewater. On the other hand, because they require intensive energy and chemicals for operation, these methods have been reported to be relatively expensive [16]. Among the various techniques of P removal, adsorption has been reported and considered to be effective and relatively stable with simple and low operation cost [3, 17]. Moreover, considering sewage sludge as an inexpensive and abundant material for adsorbent preparation, one of effective solutions is to convert it into adsorptive materials [18]. Additionally, because of their specific properties including high contents of cations (e.g., Ca^{2+}, Fe^{3+} and Al^{3+}) added during the lime-conditioned dewatering process, a large surface area, abundant functional groups that provide high quality adsorption in capturing various organic and inorganic pollutants in wastewaters, sewage sludge-based adsorbents are considered as a promising approach for treating nutrients in wastewater [19].

As an adsorbent, magnesium oxide (MgO) powder is a low-cost, highly available, non-toxic and environmental-friendly material. Because the MgO can increase water pH level (>9) and active sites of the adsorbents, it effectively enhances the removal of wastewater contaminants such as heavy metals and phosphorus and is increasingly employed for inactivating bacteria in wastewater with various HRTs from 1 to 48 hours [20–22]. By integrating magnesium oxide (MgO) into sewage sludge, we can elevate the effectiveness of wastewater treatment. The integration of MgO as a mixture with sewage sludge adsorbent can also avoid the need of chlorination and prevents the formation of harmful disinfection byproducts such as trihalomethane (THM), which are toxic to human health [23]. In addition, with its thermoplastic property, bentonite has been applied as a great binder material for improving the stability of sludge-based ceramsite adsorbents [24]. Heavy metals in the sewage sludge could be effectively stabilized through the bentonite during calcination (sintering) process [25]. Besides, zeolites, another natural material, consist of the aluminosilicate components containing exchangeable cations $\left(Na+, K^{+}, Ca^{2+}, Mg^{2+}\right.$, etc.). This specific characteristic enables zeolites to have some beneficial properties for cation exchange and adsorption processes [25].

Previously, most research studies developed and utilized a specific type of adsorbent for removing a specific sole pollutant, such as dyes, nutrients, or heavy metals ions from wastewater. For instances, low-cost materials such as algal biomass, bagasse metals, municipal sludge, basic oxygen furnace slag and waterworks sludge have been used to synthesize the adsorbents for adsorbing pollutants in wastewater such as heavy metals, dyes [26–28] and nutrients [3, 24, 25, 29]. Nevertheless, experimental studies on the simultaneous P adsorption and pathogenic activation as well as the multiple functionalities of the developed adsorbents in real applications have not been conducted.

Hence, to address the issues of P and pathogen removals from wastewater, and to achieve the proper and sustainable management of generated sludge, development of a novel sludge-based wastewater treatment technology is proposed. Hence, the objective of this study was to develop a novel multifunctional sewage-based adsorbent for simultaneously removing phosphate and pathogen from treated effluent of conventional wastewater treatment plants. Additionally, as previously discussed, because wastewater pH (depending on MgO adding) and contact times or HRT in the reactors were hypothesized to be important factors for TP and fecal coliforms removal, this research work was also conducted to evaluate insights into the significant factors affecting P and pathogens removal efficiencies by the developed adsorbents.

2. Materials and methods

2.1. Chemicals

In this study, potassium Dihydrogen Phosphate (KH_2PO_4) was used for preparing and adjusting P concentrations. Meanwhile, sodium hydroxide (NaOH) and sulfuric acid 98% (H_2SO_4) were used to prepare solutions for pH adjustment. In addition, bentonite obtained from Krungthepchemi Co., Ltd, Bangkok Thailand, zeolite obtained from Agritec & Aquaculture (Thailand) Corettd,

Bangkok, Thailand and MgO obtained from Giant Leo Intertrade Co., Ltd., Bangkok, Thailand were utilized as additive materials for synthesis of the adsorbents. The chemical compositions of the bentonite powder mainly consisted of SiO_2, Al_2O_3, Na_2O, MgO and Fe_2O_3, of 71.1%, 13.7%, 3.1%, 2.6% and 2.6%, respectively (Table 1) while the zeolite powder mainly contained SiO_2, Al_2O_3 and CaO of 66.3%, 12.9% and 3.0%, respectively (Table 2). The purity of MgO powder used in this study was 91.3% (Table 3).

Table 1. Characteristics of bentonite.

Chemicals	Compositions, %
SiO_2	71.05
Al_2O_3	13.66
CaO	0.50
MgO	2.62
K_2O	1.61
Na_2O	3.06
Fe_2O_3	2.56
FeO	0.03
TiO_2	0.14
P_2O_5	0.06

Source: Krungthepchemi Co., Ltd, Bangkok Thailand

Table 2. Characteristics of zeolite.

Chemicals	Compositions, %
SiO_2	66.26
Al_2O_3	12.97
CaO	2.95
MgO	1.08
Fe_2O_3	2.03
TiO_2	0.30
P_2O_5	0.06

Source: AAA Agritec & Aquaculture (Thailand) Co.,Ltd., Bangkok Thailand

Table 3. Characteristics of MgO powder.

Chemicals	Compositions, %
SiO_2	3.93
Al_2O_3	0.14
CaO	1.73
MgO	91.31
Fe_2O_3	0.54

Source: Giant Leo Intertrade Co., Ltd., Bangkok Thailand

2.2. Preparation of adsorbents

Overall steps of the adsorbent preparation process are presented in Fig. 1. Briefly, dewatered sewage sludge from the Nong Khaem oxidation ditch activated sludge wastewater treatment plant (WWTP) in western Bangkok, Thailand, was collected. It was oven-dried at 105°C for 24 hours, then grinded and passed through a 100-mesh sieve (ø: 150μm). The sieved sewage sludge, bentonite, zeolite and MgO powders were completely mixed with different ratios (Table 4) and manually formed into spherical granules with a diameter of $0.7 - 0.8$ cm prior to air-dried in an oven at 105°C for 24 hours [30]. After that, the dried granules were preheated at 300C for 20 minutes and then sintered at 650C for 2 hours in a muffle furnace to produce ceramsite adsorbents [31]. Five types of ceramsite adsorbents, namely SB, S30, S40, S50 and S60 (Table 5) were then selected for subsequent adsorption experiments to determine TP and fecal coliform removal.

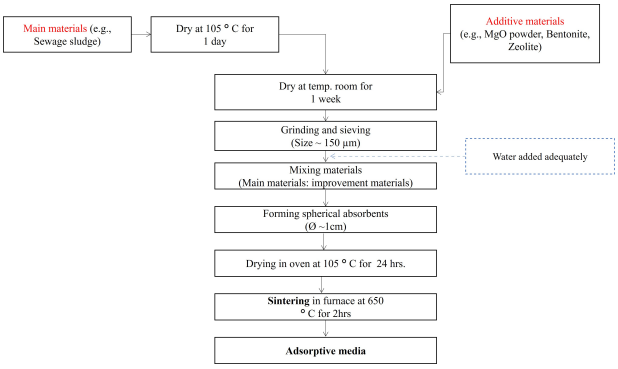

Fig. 1. Preparation of ceramsite adsorbents

2.3. Batch experiments

Synthetic wastewater with varying total phosphorus (TP) concentrations (5, 10, 25, and 50 mgP/L) was prepared by dissolving KH_2PO_4 in distilled water and adjusted initial pH to approximately 7 using 1 N diluted NaOH and H_2SO_4. Then, 15 ± 1 g of each adsorbent (SB, S30, S40, S50, and S60) and 150 mL of the synthetic wastewater were added to a 250 mL flask and mixed in a shaker at 200rpm (temperature of 25°C) for 12 hours [25]. The TP adsorption data were collected and analyzed for the adsorption isotherms [32]. Moreover, to investigate the optimum HRT for TP and fecal coliform removal, treated effluent of a wastewater treatment plant with TP and fecal coliform concentrations of approximately 11 ± 0.3mgP/L and $57,000 \pm 12,000$MPN/100 mL, respectively was used in the subsequent batch adsorption experiments with varied HRT of 1, 3, 6 and 12 hours. The duplicated tests were carried out for the batch experiments of the TP and fecal coliform removals.

2.4. Continuous filter column experiments

For continuous adsorption experiments (Fig. 2), About 1.5 kg of each adsorbent (SB, S30, S40, S50, and S60) was

Table 4. Added material ratios of the prepared sludge-based adsorbents

Adsorbent	Added material ratio%				Status
	Sludge	Bentonite	Zeolite	MgO	
SB	**50**	**50**	**0**	**0**	**Potentially stable**
S10-1	10	40	20	30	Cracked after drying
S10-2	10	50	30	10	Cracked after drying
S20-1	20	40	10	30	Cracked after stability test
S20-2	20	50	10	20	Cracked after drying
S30-1	30	30	20	20	Cracked after sintering
S30-2	**30**	**40**	**10**	**20**	**Potentially stable**
S40-1	**40**	**30**	**10**	**20**	**Potentially stable**
S40-2	40	40	5	15	Cracked after stability test
S50-1	**50**	**30**	**5**	**15**	**Potentially stable**
S50-2	50	40	5	5	Cracked after drying
S60-1	**60**	**30**	**5**	**5**	**Potentially stable**
S60-2	60	20	5	15	Cracked after sintering
S70-1	70	15	5	10	Cracked after sintering
S70-2	70	20	5	5	Cracked after sintering

Table 5. Selected ceramsite adsorbents for experiments

Adsorbent	Added material ratio%				Size (cm)	Porosity	Particle density (g/cm^3)
	Sludge	Bentonite	Zeolite	MgO			
SB	50	50	0	0	0.7	0.72	1.20
S30	30	40	10	20	0.8	0.76	2.01
S40	40	30	10	20	0.8	0.74	2.46
S50	50	30	5	15	0.8	0.77	2.07
S60	60	30	5	5	0.8	0.73	2.35

filled into a laboratory-scale column reactor with 8 cm diameter, 85 cm height, and 17 L volume. These 5 column reactors were connected with a peristaltic pump to regulate the flow rate $(0.2\,L/h)$ of influent wastewater, which came from the treated effluent of a conventional WWTP (sequencing batch reactor, SBR) at the Asian Institute of Technology (AIT) campus. The influent had TP and fecal coliform concentrations of $6.2 \pm 0.9 mgP/L$ and $24,100 \pm 8,200 MPN/100\,mL$, respectively. Based on the batch experiment results, the HRT of 6-hours was maintained in the column reactors, with continuous operation carried out for 30 days.

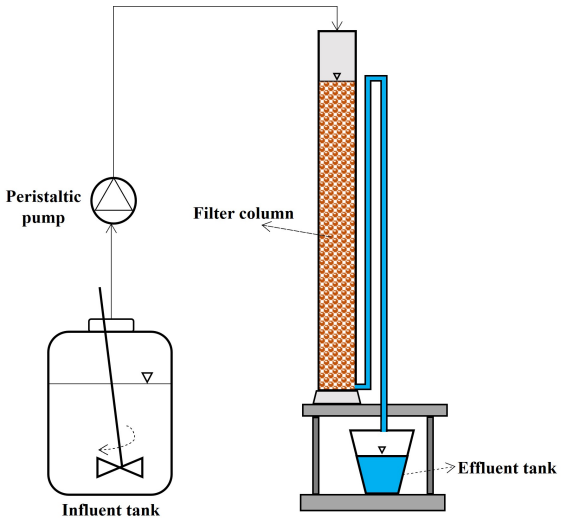

Fig. 2. Diagram of continuous filter column

2.5. Analytical methods

Influent and effluent samples were collected every 3 days and weekly for analysis of TP (Vanadate-molybdate method) and fecal coliforms (Multiple-tube fermentation method) concentrations, respectively, according to the Standard Methods for the Examination of Water and Wastewater [33]. pH values in the wastewater samples were measured using a pH meter (©Mettler Toledo AG 2009, Switzerland), respectively. The X-ray diffractometer (XRD) tech-

nique by a X-ray diffractometer (Bruker D8 ADVANCE, USA) was applied to determine the crystal structure of the adsorbents. The surface morphologies and elemental compositions of the adsorbents after adsorption were measured with the environment scanning electron microscope (ESEM) coupled with energy dispersive X-ray spectroscopy (EDS) (Thermo Fisher Scientific Quattro S, USA).

2.6. Data analysis

2.6.1. *Adsorption isotherm and kinetics for TP removal*

The adsorption capacity of TP was calculated as follows:

$$Q_e = \frac{(C_0 - C_e) \times V}{m} \quad (1)$$

$Q_e(mg/g)$ is the equilibrium adsorption capacity of phosphate, C_0 represents the initial phosphorus concentration (mgP/L), C_e is the equilibrium phosphorus concentration (mgP/L), V is the solution volume (L), and $m(g)$ is the mass of adsorbent.

Langmuir models were used to fit the experimental data, with the Eqs. (2) and (3) [32, 34]:

The Langmuir equation:

$$Q_e = \frac{Q_M K_L C_e}{1 + K_L C_e} \quad (2)$$

The linear equation can be expressed as follows:

$$\frac{C_e}{Q_e} = \frac{1}{Q_M K_L} + \frac{C_e}{Q_M} \quad (3)$$

where Q_M is the adsorbed quantity at saturation, and $K_L(L/mg)$ is Langmuir constant, respectively.

Moreover, the pseudo first-order and pseudo second-order models, expressed in Eqs. (4) and (5), respectively, were applied to describe the kinetics for TP [34].

$$\log(Q_e - Q_t) = \log Q_e - K_1 t \quad (4)$$

$$\frac{t}{Q_e} = \frac{1}{K_2 Q_e^2} + \frac{1}{Q_e} t \quad (5)$$

where, $Q_t(mg/g)$ is the adsorption capacity at the adsorption time t.K_1 $\left(h^{-1}\right)$ and K_2 (mg/g.h) are the constant rates of pseudo-first order model and pseudo-second order models, respectively.

2.6.2. *Kinetics for pathogen removal*

The kinetics for fecal coliform removal can be calculated based on first-order reaction

$$\frac{C_0}{C_e} = \mathrm{Exp}(-kt) \quad (6)$$

where C_0 represents the initial fecal coliform concentration (MPN/100mL), C_e is the final fecal coliform concentration (MPN/100mL), k is microbial removal kinetic constant $\left(d^{-1}\right)$, and t is contact time or HRT.

2.7. Statistical analysis

To statistically compare the TP treatment efficiencies and fecal coliform reductions of the different adsorbents and the column reactors, data analysis was carried out using one-way Analysis of Variance together with multiple comparison test (post-hoc test: Fisher's Least Significant Difference (LSD)) at the level of significance (α) of 0.05 in the SPSS (V.25.0). Meanwhile, the regression analysis was undertaken using Minitab (V.18.0).

3. Results and discussion

3.1. Phosphorus and pathogen removal in batch experiments

At the end of the batch experiments, the pH values in the flasks with ceramsite adsorbents SB, S30, S40, S50 and S60 were recorded as $6.8 \pm 0.1, 9.8 \pm 0.3, 9.6 \pm 0.4, 8.9 \pm 0.2$ and 8.8 ± 0.4, respectively. As shown in Figure 2, the S30, S40, S50 and S60 had better TP adsorption capacities than the SB, which had only sludge and bentonite. The TP adsorption capacities of the ceramsite adsorbents were significantly increased with increasing HRTs up to 6 hours; however, at the HRT of 12 hours, the TP adsorption capacities were not significantly increased (Fig. 3a).

The highest TP adsorption capacity $(Q_{max} = 0.45 mgP/g)$ was achieved under the highest initial TP concentration of 50mgP/L (Fig. 3b). It was also found that the initial TP concentrations had a positive effect on TP adsorption capacities $\left(R^2 > 0.9\right)$. According to Fig. 3b, the TP adsorption capacities increased with increasing initial TP concentrations $(5 - 50 mgP/L)$ at the R^2 value greater than 0.9.

Fig. 3c illustrates that the reduction of fecal coliforms by different types of ceramsite adsorbents was significantly different $(P < 0.05)$. The fecal coliform reductions were significantly increased with prolonging HRT. However, compared to the 6hour HRT data, there were no significant differences in the fecal coliform reductions at the 12 hour HRT (Fig. 3c), suggesting the HRT of 6 hours was a suitable contact time for treating TP and fecal coliforms in wastewater. The increase in pH level (> 9) could enhance the precipitation of phosphate since higher pH conditions generally promote the conversion of soluble orthophosphate ions $\left(HPO_4^{2-}\right.$ or $\left.H_2PO_4^-\right)$into less soluble forms or solid forms including calcium phosphate, iron phosphate and magnesium phosphate [29]. The previous study by Dai et al. [35] has reported the contact time of HRT of 4 hours being suitable for P adsorption and precipitation on the adsorbent surface, relatively similar to the results in this study (Fig. 3a). In addition, when the MgO powder

contact with water, it releases hydroxide ions (OH^-) that increase the pH of the solution, interfering the enzymatic activity of pathogens and disrupt their cellular functions, and ultimately leading to their inactivation [20].

3.2. Treatment performance in continuous filter column reactors

During the experiment, the average effluent pH of the SB, S30, S40, S50, and S60 reactors were $7.1 \pm 0.1, 9.7 \pm 0.6, 9.7 \pm 0.3, 8.8 \pm 0.4$, and 8.7 ± 0.48, respectively. These observed pH values were significantly lower than the pH of the zero-point charge (pHzpc) of the MgO loaded adsorbents which were previously determined to be in ranges 11.1 - 12.8 [36–38]. Accordingly, it could be anticipated that the surfaces of the ceramsite adsorbents were positively charged by MgO particles $(MgO + H_2O \rightarrow MgOH^+ + OH^-)$, resulting in an increase of the pH in wastewater [37]. In the process of phosphorus adsorption by the ceramsite adsorbents, the electrostatic interactions between the adsorbent surfaces and phosphate ions in wastewater could play an important role when the pH of the wastewater is below the [36, 37]. Due to the positive charge $(MgOH^+)$, the negatively charged phosphates such as $HPO_4{}^{2-}$ and $PO_4{}^{3-}$ were substantially adsorbed onto the adsorbent surfaces, leading to a high P adsorptive amount [36, 37] (further discussed in sections 3.3 and 3.4).

Besides, as shown in Fig. 4a, the TP removal efficiencies of the S30, S40, S50 and S60 ceramsite adsorbents were not significantly different ($p > 0.05$), being $92 \pm 4\%, 93 \pm 6\%, 92 \pm 5\%$, and $92 \pm 6\%$, but were higher than the TP removal efficiency of the SB $(85 \pm 4\%)$. This obtained results could be due to the S30, S40, S50 and S60 were added with the zeolites for improving adsorption properties and the MgO powder for increasing pH level and active sites, and hence enhancing P adsorption and surface precipitation on the adsorbents surface [36, 38]. The EDS analysis results show some amounts of TP adsorbed on the ceramsite adsorbents (Fig. 5), confirming the observed data of TP removal. It is apparent that TP removal capacity depends on the available adsorption area on the ceramsite adsorbents and would eventually decrease when the adsorption area is depleted [29]. Previous studies have reported that TP removal efficiencies of 94 to 99% can be achieved in filter column using fabricated adsorbents to treat wastewater with TP concentrations of 1-15 mgP/L at HRTs ranging from 8 to 93 hours [39–42].

Regarding fecal coliform reduction, the reductions for S30, S40, S50, S60, and SB were $3.7 \pm 0.4, 3.4 \pm 0.7, 2.6 \pm 0.5, 2.5 \pm 0.2$, and 0.2 ± 0.1 log, respectively (Fig. 4b). These results align with the pH values, with the S30 and S40 showing the high pH value (>9.5) providing more effective than the other adsorbents. The removal efficiencies of the S30, and S40 were found be greater than 99.9%. Some previous studies reported that other synthesized adsorbents such as Polyaniline/ZrO2, Lanthanum hydroxides modified poly(epichlorohydrin)-ethylenediamine composites and Magnetic porous Nickel Lanthanum- -Layered double oxides could be used to inactivate *E.* coli in wastewater between 3 - 4 log reductions at contact times ranging from 12 to 24 hours (see Table 6) [43–45]. Hence, the utilized low-cost materials (sewage sludge, zeolite and MgO powder) as well as the shorter HRT of 6 hours in this study for TP and fecal coliforms removals could make the treatment processes more cost-effective and less time-consuming.

3.3. Factors affecting treatment performance of continuous filter column reactors

In this study, the MgO powders were mixed to the adsorbents in different ratios ranging from 5% (for S60) to 20% (for S30 and S40). As shown in Fig. 6, an increase in MgO powder ratios from 0 to 20% correspondingly elevated the pH values in the reactors from 7.7 to 9.9 (Fig. 6a), sufficient for enhancing phosphorus removal by surface adsorption and precipitation [46] as well as fecal coliform inactivation [47, 48] (Fig. 6b). When applied to wastewater, MgO reacts with water, forming magnesium hydroxide $(Mg(OH)_2)$, which then dissociates into magnesium ions $\left(Mg^{2+}\right)$ and hydroxide ions (OH^-) in the wastewater [35]. The OH^- ions increase the water pH, making it more basic, which facilitated phosphorus precipitation [29]. Additionally, a higher pH can disrupt and damage pathogen cell membranes, causing proteins denaturation or loss of functional structure, leading to pathogen inactivation [48]. In addition, the amounts of TP removed were found to increase with increasing influent TP concentrations (Fig. 6c), probably because of the available adsorption area and reactive metal elements (e.g., $Mg^{3+}, Ca^{2+}, Al^{3+}$ and Fe^{3+}) of the ceramsite adsorbents for adsorption of the influent TP and the precipitated TP [29].

Although this study found the wastewater pH, initial TP concentration and HRT to have influences in efficiencies of TP and fecal coliform removal, in practice, other parameters such as initial wastewater pH, reactor type, mass of adsorbents, and temperature would also have effects on these removal efficiency data. However, because pH of treated wastewater is normally in the neutral range [49] and filter columns used in this study are commonly employed for pollutant adsorption [50], these 2 parameters were not varied in this research work. The physical-chemical reac-

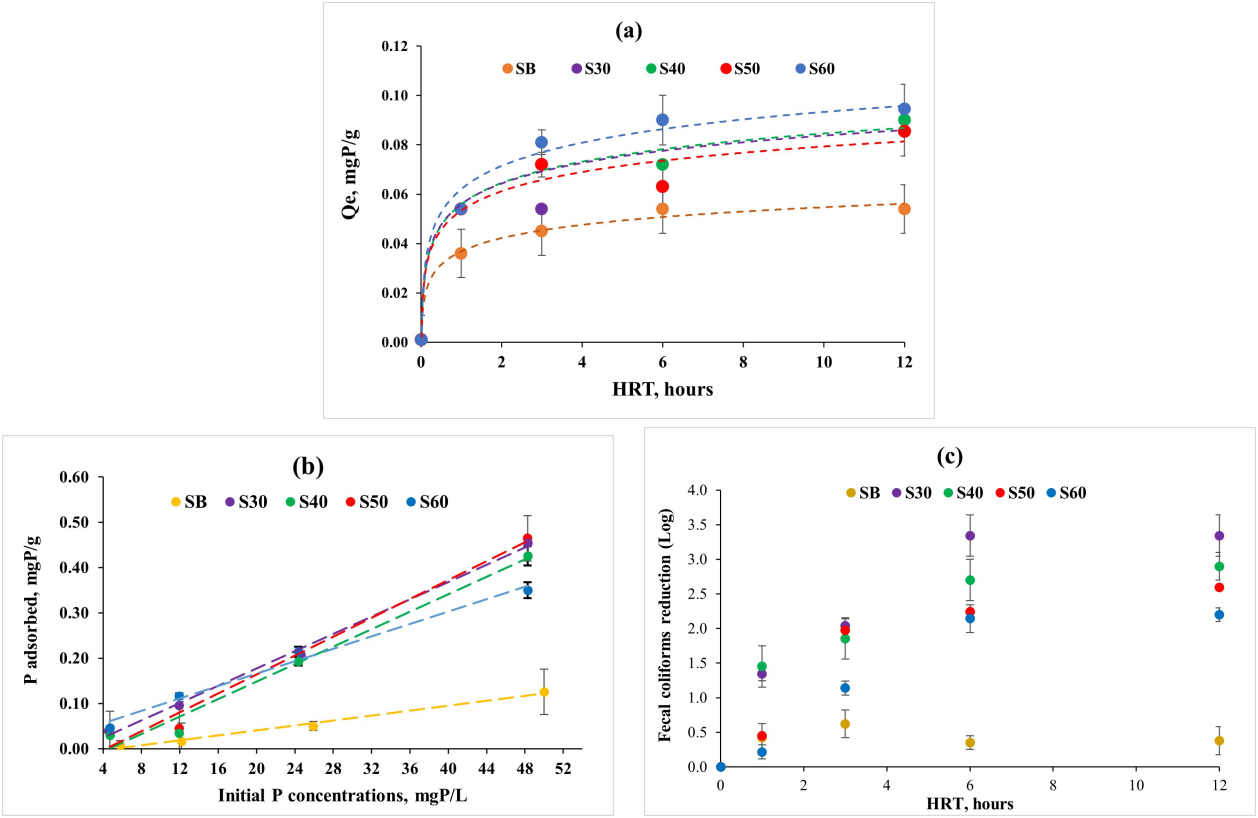

Fig. 3. TP adsorption under various HRT (a) and initial P concentrations (b) and fecal coliform reduction (c)

Table 6. Summarized comparison of treatment performance of among multifunctional adsorbents for TP and pathogenic removals

Adsorbent materials	WW	Pollutant	Condition	HRT	Treatment performance	Adsorption Isotherm	Adsorption kinetic	Refs.
Polyaniline, ZrO_2	Synthetic wastewater	TP	Adsorbent concentration: 0.002 g/L pH =6.9 (controlled) TP concentrations: 200 mgP/L	24 h	TP removal: 64% Q_{max}: 32.4 mg P/g	Langmuir	-	[43]
		E. coli	Adsorbent concentration: 0.002 g/L pH =6.9 (controlled) *E. coli*: 10^6 CFU/mL	12 h	*E. coli* removal: 99.9% (~3 log reduction)	-	-	
Lanthanum hydroxides modified poly(epichlorohydrin)-ethylenediamine composites	Synthetic wastewater	TP	Adsorbent concentration: 1.25 g/L pH = 4.0 - 10.0 TP concentrations: 5-100	24 h	Q_{max}: 186.6 mg P/g	Langmuir	Pseudo second-order kinetic	[44]
		E. coli	Adsorbent concentration: 1.25 g/L pH = 4.0 - 10.0 *E. coli*: 10^4 CFU/mL HRT:		*E. coli* removal: 100 % (~ 4 log reduction)	-	-	
Magnetic porous NiLa-Layered double oxides	Secondary effluent	TP	Adsorbent concentration: 1.25 g/L pH = 5.5–8.5 TP concentrations: 0- 400m mgP/L	24 h	Q_{max}: 203.10 mgP/ g	Langmuir	Pseudo second-order kinetic	[45]
		E. coli	Adsorbent concentration: 1.25 g/L pH = 5.5–8.5 *E. coli*: 10^4 CFU/mL		*E. coli* removal: 99.99% (~ 4 log reduction)	-	-	
Sewage sludge, zeolite, MgO	Secondary effluent (adjusted)	TP	Adsorbent concentration: 0.1 g/L pH: 7.1 - 9.8 (final pH) TP concentration: 2-50 mgP/L	6 h	TP removal: 93% (initial TP: 6 mg/L) Q_{max}: 0.45 mgP/g	Langmuir	Pseudo-second-order	This study
		Fecal coliforms	*Fecal coliforms:* $2.4x10^4$ MPN/100mL		*Fecal coliforms* removal 99.99% (~ 3.7 log reduction)	-	First-order model	

tions occurring in the filter column could be also affected by temperature, but this study was conducted under ambient tropical conditions (average temperature $\sim$ 30°C) to simulate actual operations of the wastewater treatment

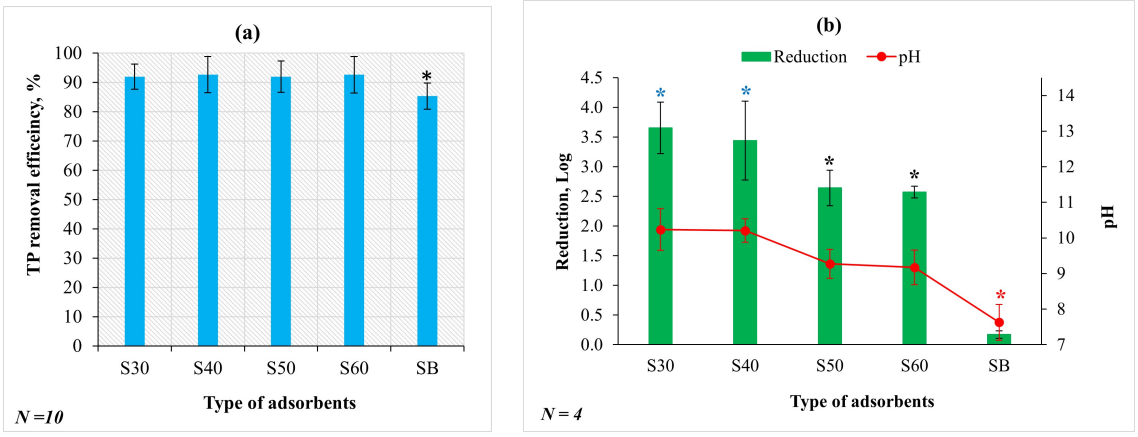

Fig. 4. TP removal (a) and fecal coliform reduction (b) in filter column reactors (Note: * (different colors) the mean differences are significant at the 0.05 level; see further results of post-hoc test in Tables 7 and 8, respectively)

Table 7. Multiple comparisons for TP removal

Post Hoc Tests: Fisher's Least Significant Difference (LSD)

		Mean Difference			95% Confidence Interval	
(I) Media	(J) Media	(I-J)	Std. Error	Sig.	Lower Bound	Upper Bound
S30	S40	-.76200	1.86900	.688	-4.6607	3.1367
	S50	-.03200	1.86900	.987	-3.9307	3.8667
	S60	-.72200	1.86900	.703	-4.6207	3.1767
	SB	6.56800*	1.86900	.002	2.6693	10.4667
S40	S30	.76200	1.86900	.688	-3.1367	4.6607
	S50	.73000	1.86900	.700	-3.1687	4.6287
	S60	.04000	1.86900	.983	-3.8587	3.9387
	SB	7.33000*	1.86900	.001	3.4313	11.2287
S50	S30	.03200	1.86900	.987	-3.8667	3.9307
	S40	-.73000	1.86900	.700	-4.6287	3.1687
	S60	-.69000	1.86900	.716	-4.5887	3.2087
	SB	6.60000*	1.86900	.002	2.7013	10.4987
S60	S30	.72200	1.86900	.703	-3.1767	4.6207
	S40	-.04000	1.86900	.983	-3.9387	3.8587
	S50	.69000	1.86900	.716	-3.2087	4.5887
	SB	7.29000*	1.86900	.001	3.3913	11.1887
SB	S30	-6.56800*	1.86900	.002	-10.4667	-2.6693
	S40	-7.33000*	1.86900	.001	-11.2287	-3.4313
	S50	-6.60000*	1.86900	.002	-10.4987	-2.7013
	S60	-7.29000*	1.86900	.001	-11.1887	-3.3913

*. The mean difference is significant at the 0.05 level.

systems, which to save costs, normally do not increase their wastewater temperatures by using external energy. Furthermore, since mass of adsorbents could have effects on the specific surface areas for adsorption of TP and fecal coliforms, further research for evaluate the effect of this parameter on wastewater treatment efficiency is strongly recommended.

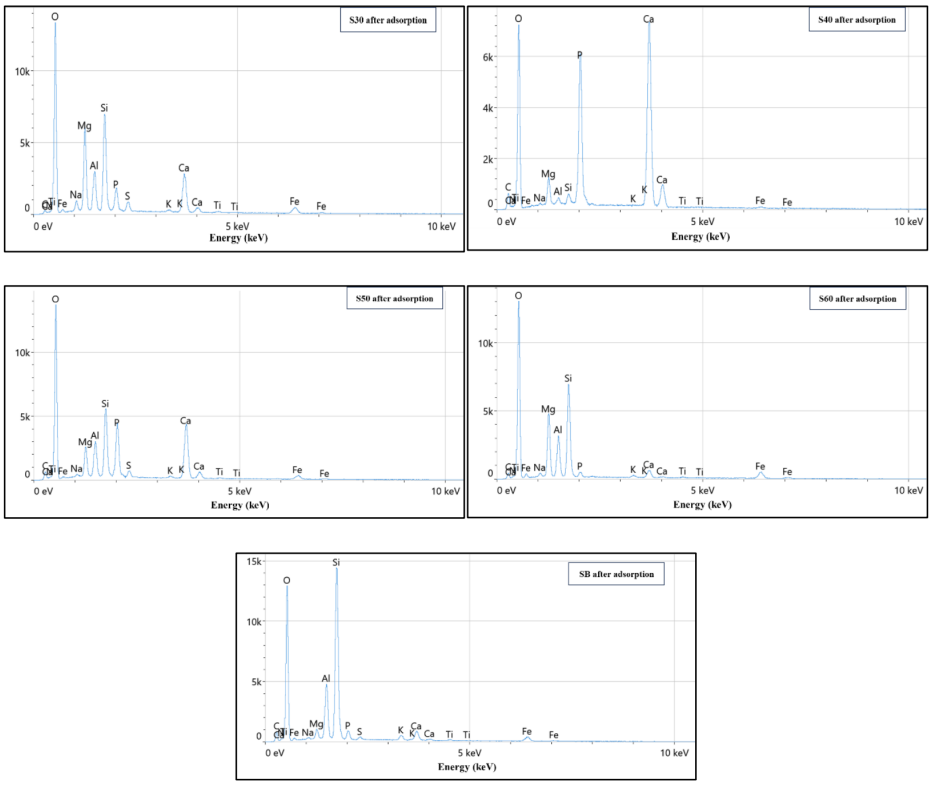

Fig. 5. EDS analysis of adsorbents

3.4. Characterizations of adsorbents

Further insights into the TP removal process by the adsorbents were investigated from the analysis of mineralogical compositions, morphological properties, chemical compositions by XRD, ESEM, and EDS analyses, respectively (Figs. 5, 7 and 8, respectively). According to the obtained results of the XRD analysis, Ca and Mg - containing phases such as calcite ($CaCO_3$), Periclase (Fe_2MgO_4) were mainly detected in the S30, S40, S50 and S60, corresponding to the chemical compositions of the added materials, namely bentonite, zeolite, and MgO powders (Tables 1 to 3). Specially, the XRD analysis of the S40 indicated that the adsorbents contained a variety of crystal phases including mainly Quartz (SiO_2) , Calcite ($CaCO_3$), Periclase (Fe_2MgO_4) (Fig. 9a).

During the sintering processes, when the temperature was raised to 650°C, the volatile components were evaporated, and microstructure and mineral phases could be completely formed in the synthesized ceramsite adsorbents [31]. The SiO_2 and $CaCO_3$ crystals were reported to improve the strength of ceramsite adsorbents, while other active components such as Ca^{2+}, Mg^{3+}, and Fe^{3+} could enhance phosphorus precipitation [29, 51, 52]. Obviously, EDS mapping demonstrated TP accumulation via adsorption and precipitation on the adsorbent surface (Fig. 9b). Besides, the EDS analysis showed Mg, Ca, O, P, and Al as the main elements forming the precipitates, further confirming the high TP amounts precipitated and adsorbed on the surface of the developed adsorbent (Fig. 9c), The TP adsorption by the ceramsite adsorbents can be achieved by firstly adsorbing the P in form of phosphate $(PO_4{}^{3-})$ from wastewater onto the surface of the ceramsite adsorbents and then removing phosphorus by chemical adsorption (ion exchange) and solid surface deposition throughout chemical bonds and electrostatic attraction [46]. At high pH values (> 9), the PO_4^{3-} in the wastewater was adsorbed and then reacted with the Mg^{3+}, Ca^{2+}, Al^{3+}, Fe^{3+} in the ceramsite adsorbents to form phosphorus precipitates, according to Eq. (7) [46]:

$$xPO_4{}^{3-} + 3M^{x+} = M_3\,(PO_4)\,x\downarrow \tag{7}$$

where M represents metal elements Mg, Ca, Al, Fe and etc.

Besides, since secondary effluent and surface water typically contain a variety of anions (e.g., $HCO_3{}^-$, $SO_4{}^{-2}$, Cl^-, $NO_3{}^-$) and cations (e.g., $K^+$and Na^+) and dissolved organic matters (fulvic acid (FA) and

Table 8. Multiple comparisons for fecal coliform reduction

Post Hoc Tests: Fisher's Least Significant Difference (LSD)

		Mean Difference			95% Confidence Interval	
(I) Media	(J) Media	(I-J)	Std. Error	Sig.	Lower Bound	Upper Bound
S30	S40	.21800	.48344	.657	-.7904	1.2264
	S50	1.01600*	.48344	.048	.0076	2.0244
	S60	1.08600*	.48344	.036	.0776	2.0944
	SB	3.48600*	.48344	.000	2.4776	4.4944
S40	S30	-.21800	.48344	.657	-1.2264	.7904
	S50	.79800	.48344	.114	-.2104	1.8064
	S60	.86800	.48344	.088	-.1404	1.8764
	SB	3.26800*	.48344	.000	2.2596	4.2764
S50	S30	-1.01600*	.48344	.048	-2.0244	-.0076
	S40	-.79800	.48344	.114	-1.8064	.2104
	S60	.07000	.48344	.886	-.9384	1.0784
	SB	2.47000*	.48344	.000	1.4616	3.4784
S60	S30	-1.08600*	.48344	.036	-2.0944	-.0776
	S40	-.86800	.48344	.088	-1.8764	.1404
	S50	-.07000	.48344	.886	-1.0784	.9384
	SB	2.40000*	.48344	.000	1.3916	3.4084
SB	S30	-3.48600*	.48344	.000	-4.4944	-2.4776
	S40	-3.26800*	.48344	.000	-4.2764	-2.2596
	S50	-2.47000*	.48344	.000	-3.4784	-1.4616
	S60	-2.40000*	.48344	.000	-3.4084	-1.3916

*. The mean difference is significant at the 0.05 level.

humic acid (HA)), which are anticipated as interferences for the P adsorption [36, 38, 53], it is crucial to examine adsorption selectivity of the ceramsite adsorbents to explain how those ions affect the P adsorption process. There were several experiments conducted to investigate the adsorption selectivity of the P adsorption of the MgO loaded adsorbents [36–38, 53]. The results of these experiments correspondingly confirmed that due to their high selectivity for adsorbing the phosphate ion, the ions including Cl^{-}, $NO_3{}^{-}$and $SO_4{}^{2-}$ and the FA and HA had negligible effects on the P adsorption by the MgO loaded adsorbents [36–38, 53], whereas the presence of $HCO_3{}^{-}$could influence the phosphorus adsorption by the MgO loaded adsorbents [36–38]. This can be explained by the fact that at pH above 8.5 , the $HCO_3{}^{-}$ionizes to produce $CO_3{}^{2-}$, which shares similar the Lewis basicity with phosphate; consequently, the produced $CO_3{}^{2-}$ could compete with the phosphate ions for available active sites, affecting the overall phosphorus adsorption process [36, 37].

Additionally, the production and treatment costs of the ceramsite adsorbent (S40) in this study were estimated to be $\sim$ 0.74US$/kg and $\sim$ 1.67US$/$m^3$, respectively (see Tables 9 and 10), which were relatively lower than that of conventional adsorbents in the global market have been reported between $0.8 - 5$US$/kg and 50-200 US $/$m^3$, respectively [54, 55]. Similarly, when compared to the treatment costs of other previously developed adsorbents such as chitosan-calcite and nanoscale zero-valent iron adsorbents, being $\sim$ 3 and $\sim$ 4 US $/$m^3$, respectively [55, 56], the treatment cost of the adsorbent in this study was found to be lower, suggesting the ceramsite adsorbent was more cost-effective for wastewater treatment. Nevertheless, because this information is in the preliminary stage for in determining the costeffectiveness, a further study on the techno-economics or feasibility of using the ceramsite adsorbents for wastewater treatment in upscaled or pilot-scale application are recommended. Furthermore, the obtained results of TP adsorbed onto the adsorbents also suggested the potential of the developed adsorbents used for simultaneous removal and recovery of the TP from the wastewater. Considering the concept of the circular economy and cost-

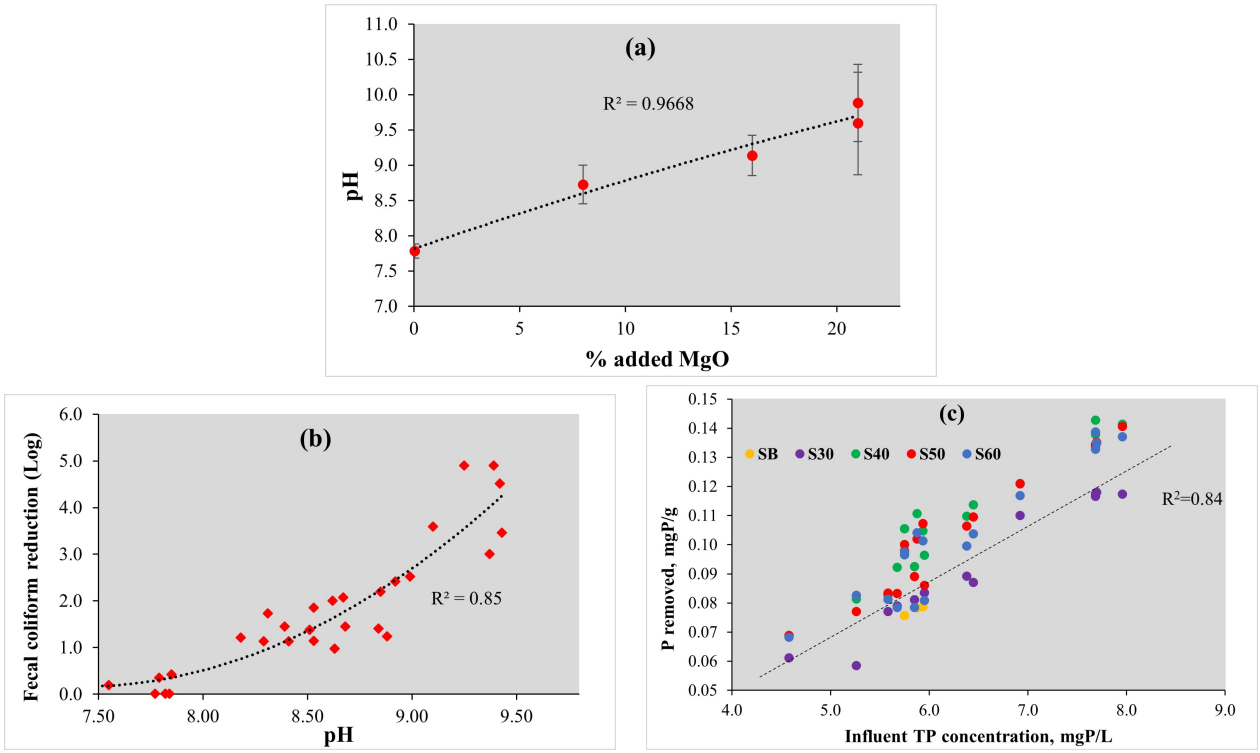

Fig. 6. Correlations of added MgO powder and pH (a) and pH and fecal coliform reduction (b) and Influent TP and removed TP (c)

effectiveness, instead of regenerating the used adsorbents by either chemical or thermal desorption process, this end-product, P-enriched adsorbents could be directly reused as fertilizer for agriculture purpose [57], potentially offering a more economical alternative to commercial fertilizers since they should be cheaper than commercial fertilizers.

Table 9. Estimated cost production of ceramsite adsorbent (S40).

Cost components	Cost (**US$/kg**)*
Prepared sludge	0.01
Zeolite	0.03
Bentonite	0.02
MgO	0.02
Water	0.01
Molding (forming)	0.05
Power consumption in sintering process	0.59
Total	**0.74**

*The production cost (US$) per unit of the adsorbent mass (kg) was estimated based on the costs of the used raw materials (commercial prices) and production processes (industrial scale) (Ighalo et al., 2022).

3.5. Adsorption Isotherms and kinetics

3.5.1. Adsorption isotherm for TP removal

For adsorption isotherms of the ceramsite adsorbents, both Langmuir and Freundlich models were used for evaluating the adsorption isotherms [32, 34, 58]. However, only the Langmuir model was found to fit with the experimental data, whereas the obtained data did not fit well with the Freundlich model. The Langmuir model fitting for the batch experiments of the S30, S40, S50, and S60 adsorbents for TP adsorption are shown in Table 5 (also see Fig. 10). The SB50 and SB60 adsorbents with pH of 8.9 and 8.8, respectively, had the R^2 values of 0.94 and 0.98 , respectively. However, the data of TP adsorption for the S30 and S40 adsorbents with pH of 9.8 and 9.6, respectively, had low R^2 values. The results suggest that the TP removal by the S50 and S60 adsorbents was probably driven by the process of homogeneous and monolayer adsorption [23]. Due to relatively high pH values, the removal of TP by S30 and S40might involve multiple mechanisms, including adsorption to metal oxide/hydroxide (specifically MgO), electrostatic attraction, surface complexation, and TP precipitation. on both solid surface and in liquid phase [29], thus contributing to the overall TP removal. This multifaced removal process could account for the imperfect fit of S30 and S40 data with

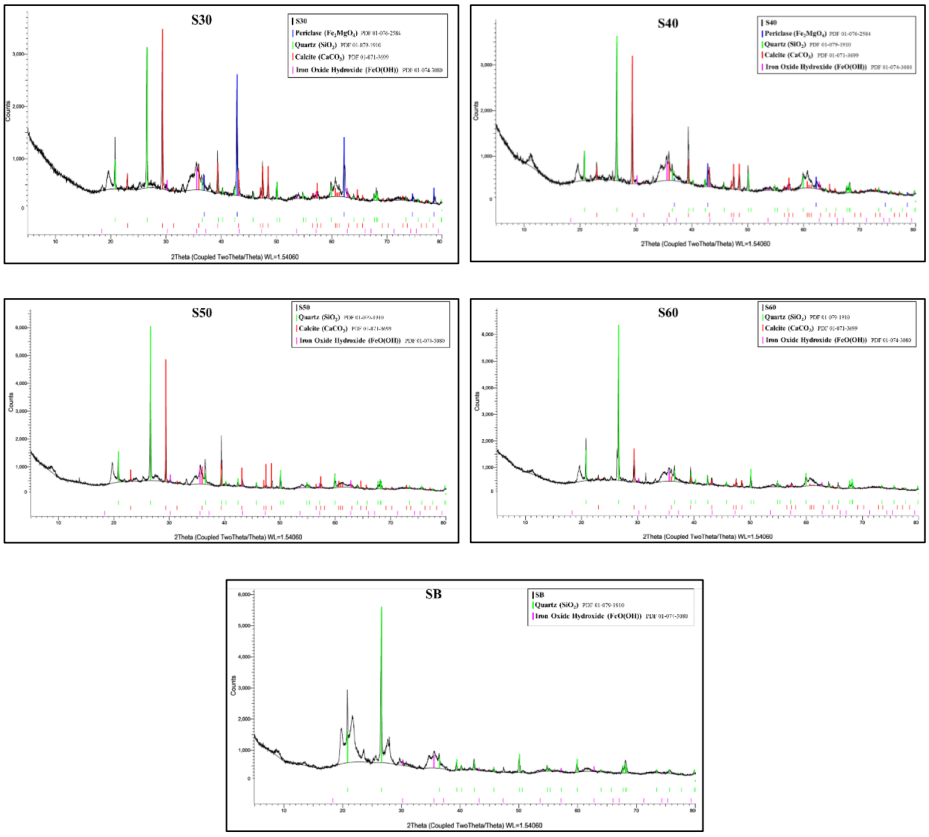

Fig. 7. XRD analysis of adsorbents

the Langmuir model, which assumes homogeneous and monolayer adsorption.

3.5.2. *Adsorption kinetics for TP removal*

The pseudo-first- and pseudo-second-order kinetic models for TP removal by the developed ceramsite adsorbents are revealed in Figs. 11 and 12, respectively, with TP adsorption kinetic model parameters presented in Table 11. The TP adsorption by the SB, S30, S40, S50 and S60 adsorbents aligned better with the pseudo-second-order kinetics (R^2 values of 0.99 , 0.97, 0.98, 0.97, 0.99) than with the pseudo-first-order kinetic model (R^2 values of 0.73, 0.77, 0.87, 0.83, 0.86). Hence, this indicates that the pseudo-second-order kinetics provided a more accurate description of the TP removal process than the pseudo-first-order kinetics. Factor influencing TP adsorption capacities and efficiencies possibly include effective adsorption sites, initial TP concentration, adsorption rates, contact times, and HRTs. These findings were also correspondingly similar to those reported by previous studies [29, 32, 48, 59].

3.5.3. *Kinetics of fecal coliform removal*

In this study, the first-order model could describe well the kinetics of the fecal coliform removal in the ceramsite adsorbents, namely S30, S40, S50 and S60 (R^2 values: 0.88, 0.98, 0.88 and 0.89, respectively, as shown in Fig. 13). The kinetic constants for bacterial decay were 0.36, 0.29, 0.26 and 0.25 d^{-1} for the S30, S40, S50 and S60 adsorbents, respectively, correlating with the mixed MgO powder ratios and pH data. This suggests the potential of the developed ceramsite adsorbents in fecal coliform removal, serving as a disinfection unit and reducing the use of chlorine, which produces toxic by-products such as trihalomethane (THM) [23], or other disinfectants such as ozone or ultraviolet light which are generally more expensive in investment, operation and maintenance costs.

4. Conclusions

The novel multifunctional ceramsite adsorbents were developed using sewage sludge mixed with different ratios of the MgO powder to simultaneously removing TP and fecal coliform in the treated effluent wastewater. At the optimum HRT of 6 hours, the developed ceramsite adsorbents

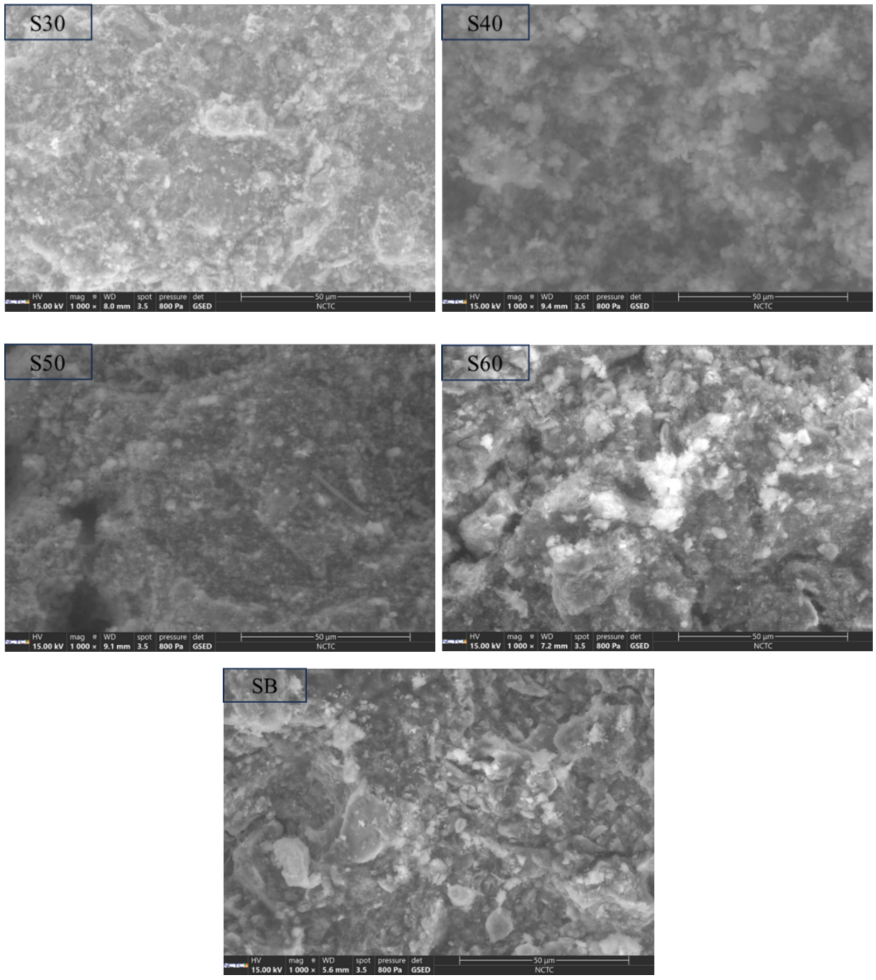

Fig. 8. ESEM analysis (1000X) of adsorbents

could effectively remove TP (97%; 0.14mgP/g) and inactivate fecal coliforms (3.4 log reduction) in the wastewater. The ratios of added MgO powder increased pH values to be greater than 9, resulting in enhancing TP and fecal coliform removal in the filter column reactors. The developed ceramsite adsorbents as a novel adsorptive material in this study present a promising solution to treat wastewater contaminated with phosphorus and pathogens, addressing the pressing need for more sustainable wastewater treatment methods. However, evaluation of the techno-economics or feasibility of developed ceramsite adsorbents in upscaled wastewater treatment units are recommended.

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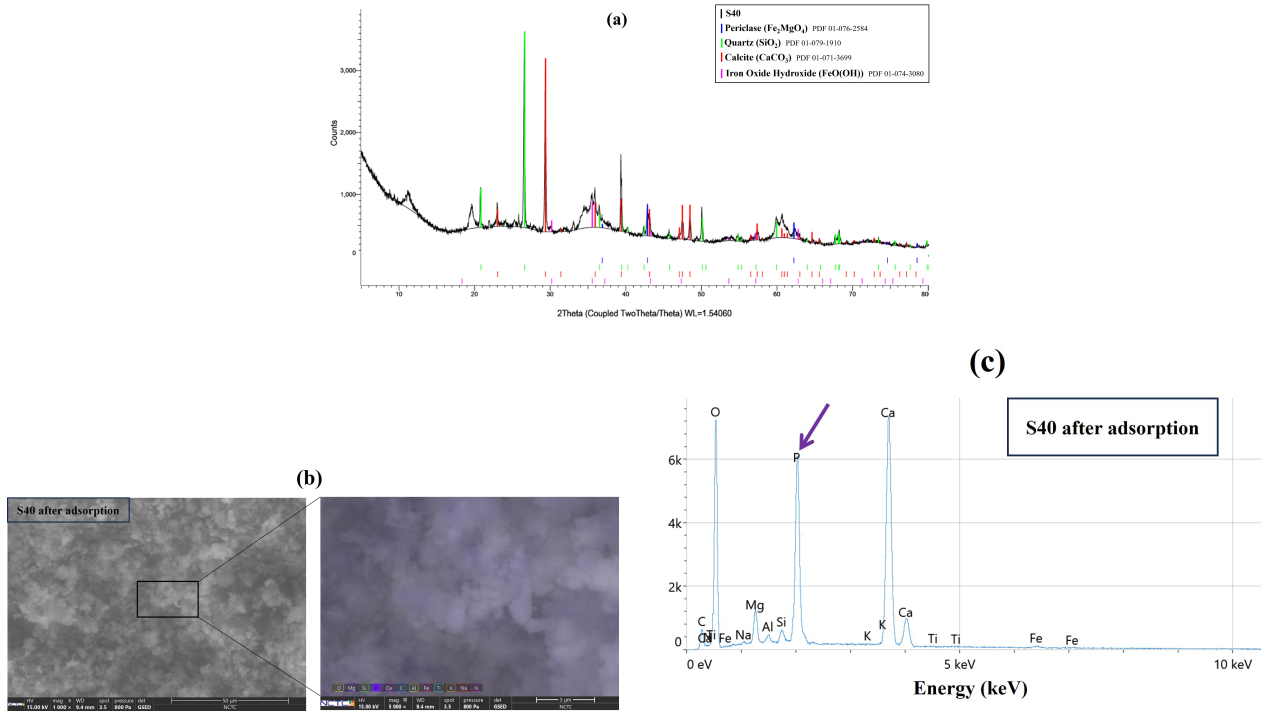

Fig. 9. XRD (a) SEM (b) and EDS (c) analysis of adsorbent (S40) and phosphorus removal mechanisms by ceramsite adsorbents (d)

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Table 10. costs of different adsorbents.

Adsorbents	Production costs ($\mathbf{US\$kg}$)	Treatment cost ($\mathbf{US\$m^3}$)	Refs.
Ceramsite adsorbent (S40)	0.74	1.67*	This study
Chitosan-calcite adsorbent	0.56	4.00	[55]
Nanoscale zero-valent iron adsorbent	-	3.15	[56]
Commercial activated carbon (AC)	$0.8-5$	$50-200$US\$ m^3	[54, 55]

*The treatment cost was calculated according to the method by Ighalo et al. (2022).

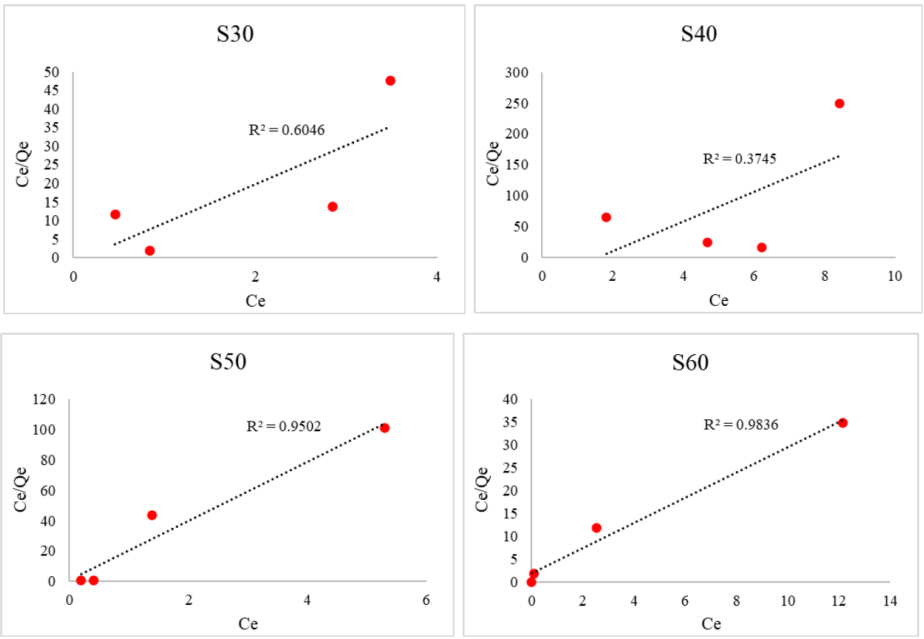

Fig. 10. Langmuir linear adsorption isotherm

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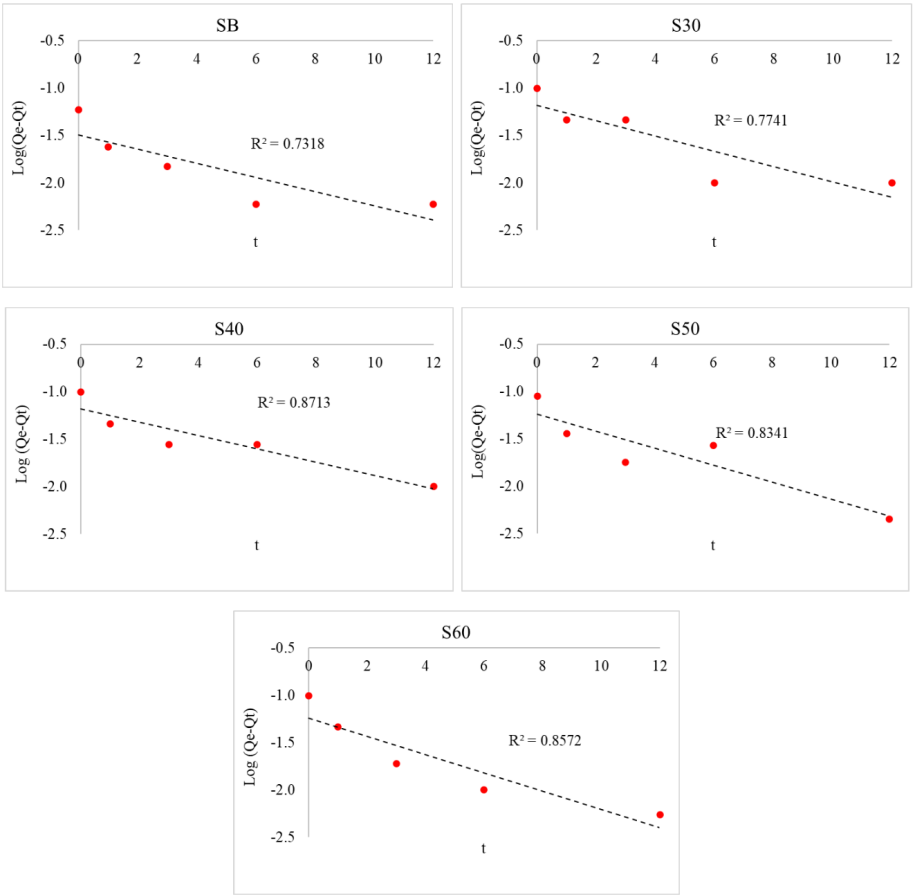

Fig. 11. Pseudo-first-order kinetic model for TP adsorption

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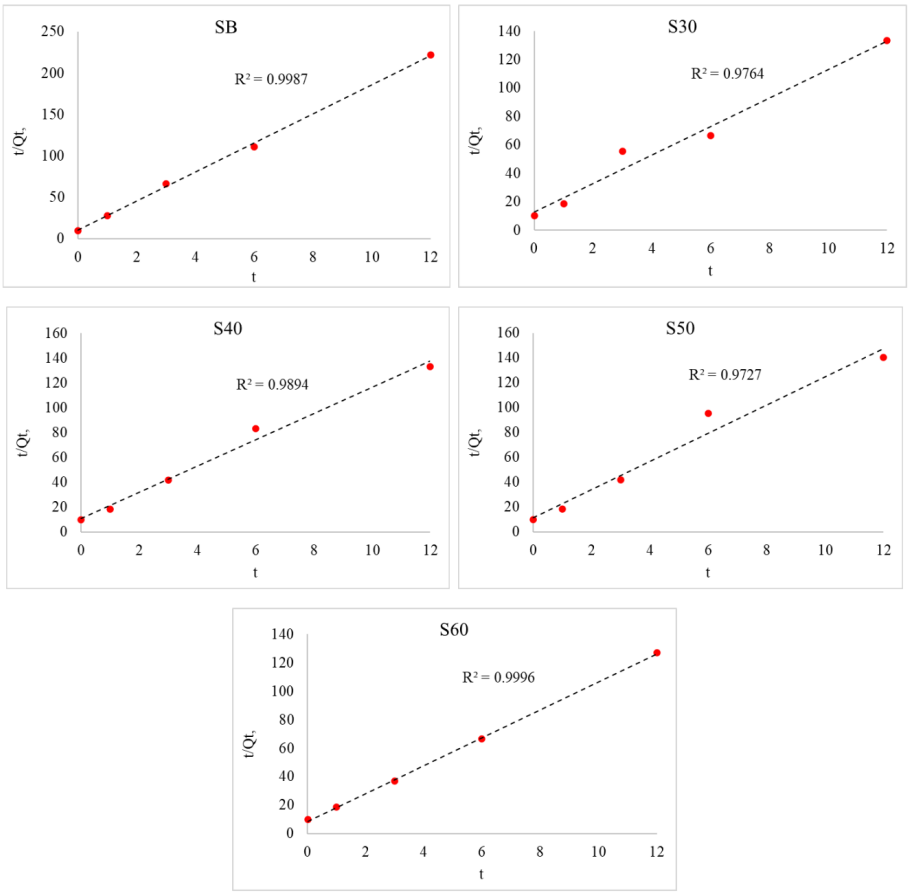

Fig. 12. Pseudo-first-order kinetic model for TP adsorptions

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Table 11. Parameters of isotherm and kinetics for TP removal. Note: - Data are not following the model.

Models	Parameters	Adsorbents				
		SB	S30	S40	S50	S60
Langmuir	Q_e (mg P/g)	-	0.096	0.041	0.051	0.370
	K_L (L/mg)	-	8.43	0.62	19.00	1.41
	R^2	-	0.60	0.37	0.95	0.98
Pseudo-first-order	Q_e(mg P/g)	0.03	0.07	0.07	0.06	0.06
	$K_1(h^{-1})$	0.074	0.081	0.070	0.089	0.096
	R^2	0.73	0.77	0.87	0.83	0.86
Pseudo-second- order	Q_e (mg P/g)	0.06	0.10	0.09	0.09	0.10
	K_2 (mg P/g.h)	29.9	7.9	10.1	11.3	11.0
	R^2	0.99	0.97	0.98	0.97	0.99

Note: - Data are not following the model.

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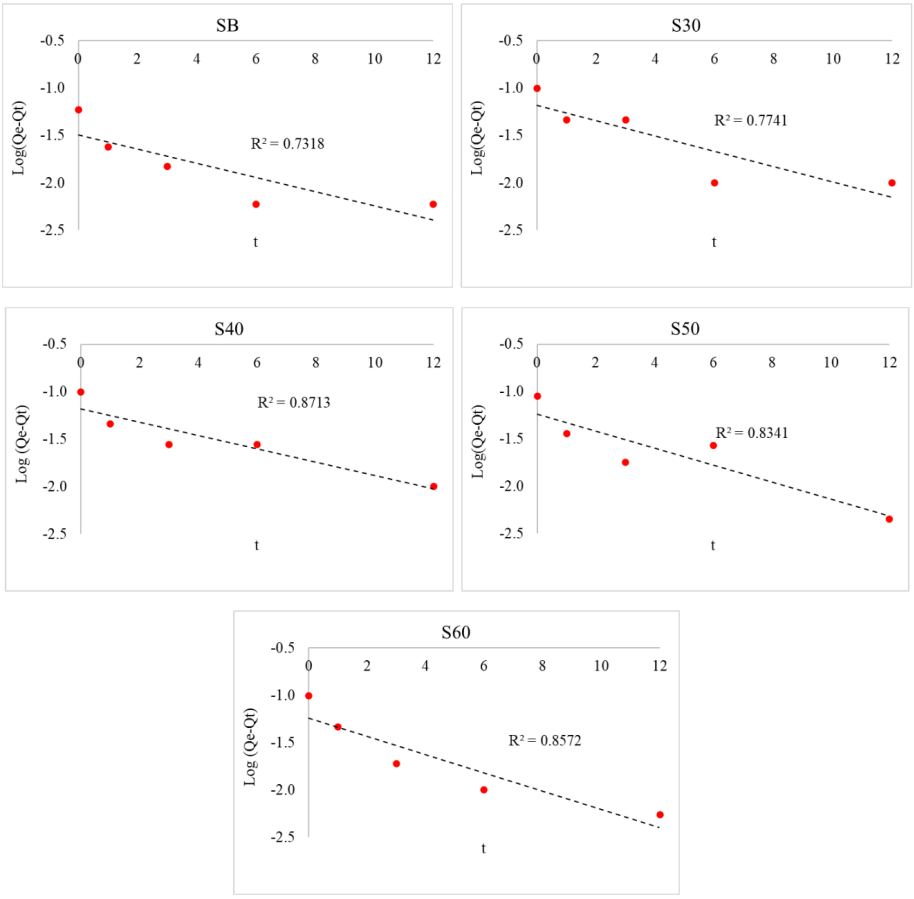

Fig. 13. First order kinetics of fecal coliform removal

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