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ARTICLE INFO Article History: Received 14 September 2021 Revised 9 November 2021 Accepted 22 November 2021 Available online 12 December 2021 Keywords: eutrophication phosphate removal thiamine magnetic iron oxide adsorption plant growth ABSTRACT Background: The pressing demand to increase agricultural productivity amid the rapidly growing population has exponentially boosted fertilizers usage. Phosphate (Pi) runoff from fertilizers induces eutrophication in water sources and severely affects its surrounding ecosystems. To cope with Pi accumulation problem, this study

**1** reported the synthesis of an environmentally friendly magnetic adsorbent

, namely Fe<sub>3</sub>O<sub>4</sub>/thiamine (thF). Method:

**1** A one-step chemical oxidation and functionalization technique for thF synthesis was

developed. X-ray diffraction (XRD), X-ray photoelectron spectroscopy (XPS), Fourier-transform infrared spectroscopy (FTIR), nitrogen (N<sub>2</sub>) sorption,

**22and superconducting quantum interference device (SQUID) analysis were**  
con- ducted **to ensure the**

formation of Fe<sub>3</sub>O<sub>4</sub>, confirm the successful incorporation of thiamine, and gain insight into the factors influencing the adsorptivity of thF-363. Significant Findings: The thF synthesized at 363 K (thF-363) produces an adsorbent with the highest Pi removal efficiency compared to other synthesis conditions. The thF-363 showed

**1up to 1.51-fold higher adsorption capacity than the**

unmodified Fe<sub>3</sub>O<sub>4</sub>. The large surface area and occurrence of thiamine functional groups are the contributing factors in enhancing its adsorption capacity for Pi removal. The thF-363 did not adversely affect the growth of the model plant, *Arabidopsis thaliana*; demonstrating its suitability as an environmentally friendly adsorbents for Pi removal from eutrophicated water with the feasibility of magnetic separation from an aqueous system. © 2021 Taiwan Institute of Chemical Engineers. Published by Elsevier B.V. All rights reserved. 1. Introduction Phosphorus in phosphate (Pi) form is essential for every organism, including humans, animals, and plants. Appropriate Pi intake is majorly related to bone?teeth health and overall growth for animals and humans [1]. Similarly, Pi availability is also affiliated to plant vigor, where Pi deficiency had been acknowledged to affect root growth. The prolonged starvation eventually can be detrimental to the sustainability of agriculture [2,3]. Due to its indispensable nature, \* Corresponding Author: Artik Elisa Angkawijaya,

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E-mail addresses: artikelisa@mail.ntust.edu.tw, artikelisa@yahoo.com (A.E. Angkawijaya). y These authors contribute equally. most global Pi consumption is allocated for the production of fertil- izers and animal feed supplements [4,5]. Pi-fertilizers are regularly applied on agricultural soil to maintain adequate-high levels of Pi to satisfy the plant demand for growth and development. However, only less than half of the administered Pi is absorbed by crops while the remainder is leached to the environment through soil and often end-up in water [6,7]. Accumulation of Pi in the water body had been acknowledged as the primary cause of eutrophication which sets off a series of detrimental effects in the aquatic ecosystem; from the algal bloom to the generation of hypoxic or anoxic 'dead zone' which affect the availability of oxygen in water [8]. To alleviate this issue, various methods such as chemical precipitation, biological treatment, anion exchange, electro-coagulation, acid-thermal treat- ment, and adsorption have been developed to efficiently remove <https://doi.org/10.1016/j.jtice.2021.104162> 1876-1070/© 2021 Taiwan Institute of Chemical Engineers. Published by Elsevier B.V. All rights reserved. excess Pi from water resources [9-16]. Among them, adsorption is one of the most used methods in managing Pi concentration in water, owing to its inexpensive and straightforward operation. Several composite adsorbents prepared from a combination of inorganic and organic compounds had been widely used for Pi removal application. For instance, MgO/biochar possessed Pi adsorp- tion capacity up to 122 mg/g [17], the adsorption capacity of mont- morillonite-iron crosslinked alginate, and its zirconium modified alginate can reach 48.78 mg/g and 67.72 mg/g, respectively [18,19]. While various adsorbents with excellent adsorption capacity for Pi removal had been developed, separation

of the post-adsorption adsorbent from the water is the main issue that leads to the impracticability of those adsorbents. Filtration and/or centrifugation procedure are commonly performed to recover the adsorbent from treated water. In this work, an adsorbent with magnetic-separability property was developed, with Fe<sub>3</sub>O<sub>4</sub> as the core material. Superparamagnetic Fe<sub>3</sub>O<sub>4</sub> possesses remarkable magnetic properties that allow its practical separation post-adsorption using an external magnetic field [20-22]. Besides their superparamagnetic and adsorbing properties, the low toxicity, durability, biocompatibility, low cost, and chemical stability of Fe<sub>3</sub>O<sub>4</sub> made them the promising adsorbent for water treatment [23-26]. In this study, a composite material consisting of Fe<sub>3</sub>O<sub>4</sub> and thiamine was developed to generate magnetic adsorbent with enhanced adsorption capacity. Thiamine, also known as Vitamin B<sub>1</sub>, is a water-soluble vitamin beneficial for plants and animals due to its antioxidant activity. It may also function as a cofactor for various metabolic activities [27,28]. In plants, thiamine is distributed in leaves, seeds, flowers, and roots; and is known to play a role to protect plant from abiotic and biotic stresses [29,30]. Thiamine molecule consists of

**18a pyrimidine ring (4-amino-2-methyl-5-pyrimidyl)** which is connected **to a thiazole ring (4-methyl-5-b-hydroxyethylthiazolium)** through **a methylene bridge**

[31]. The occurrence of these N-functional groups in thiamine, which tends to be positively charged, is expected to promote the adsorption capacity toward anionic Pi ions [32-34]. In addition, through the beneficial effect of thiamine for plant stress adaptation, the post-adsorption thiamine-functionalized Fe<sub>3</sub>O<sub>4</sub> (thF) can be a potential Pi-supplement to support plant growth. This study also

**24 investigated the effect of synthesis temperature on the product yield and its Pi-removal**

performance. Several physicochemical characterizations were carried out to elucidate the properties of thF, including crystallinity pattern, surface chemistry, surface functional groups, porosity, and magnetic behavior. The physicochemical properties of thF were compared to that of unmodified Fe<sub>3</sub>O<sub>4</sub> (F) to confirm the successful incorporation of thiamine. Adsorption isotherm of Pi by thF and F at different temperatures was conducted to study their adsorption behavior and mechanism. The environmental compatibility of these adsorbents was also assessed.

**292. Materials and methods 2.1. Materials All chemicals used were of analytical grade and were used**

without any pretreatment. Iron(II) sulfate heptahydrate (FeSO<sub>4</sub>·7H<sub>2</sub>O, purity >99.5%) and hydrochloric acid (HCl, purity 37%) were purchased from Acros Organics, New Jersey, USA. Ammonia (NH<sub>3</sub>, purity 28-30%) and potassium dihydrogen phosphate (KH<sub>2</sub>PO<sub>4</sub>, purity 99.5%) were manufactured by Showa, Tokyo, Japan. Thiamine hydrochloride (C<sub>12</sub>H<sub>17</sub>CIN<sub>4</sub>OS.HCl, purity 99%) was a product of Sigma, St.Louis, MO, Germany.

**2Sodium hydroxide (NaOH, purity >97%) was obtained from**

Fisher Scientific, Loughborough, UK. The

**2solutions used for the experiments were freshly prepared**

before use by dissolving a certain amount of chemicals in deionized (DI) water with 18.3 MV/cm. 2.2. Synthesis of magnetite-based adsorbent For thF synthesis, 0.2024 g of thiamine and 0.1668 g of FeSO<sub>4</sub>·7H<sub>2</sub>O (Fe salt)

**25 were dissolved in 20 mL of DI water. The solution was**

left to react for 1 h under constant mixing at 300 rpm at certain temperatures, as listed in Table 1. Subsequently, ammonia was added (

**1 molar ratio of NH<sub>4</sub>OH, Fe salt, and thiamine 10:1:1). The**

mixture was then left to react for another hour. The precipitate generated from the reaction was magnetically separated from the supernatant with an external magnetic force. The pH of the supernatants was determined by Denver Instrument UltraBasic pH Benchtop Meters UB-10 and was presented as the solution pH at the end of the reaction (Table 1). On the other hand, the solid product was washed at least 3

**16 times with DI water and ethanol and dried at 313 K under vacuum for 24 h. The**

unmodified Fe<sub>3</sub>O<sub>4</sub> was prepared using a similar procedure, without the addition of thiamine, and used as a control. 2.3. Characterization

**1 Thermogravimetric analysis (TGA) was performed using a TA instruments/TGA 550, operated at a temperature range of 303-973K with a heating rate of 10 K/min under N<sub>2</sub> flow. The**

crystal pattern of the prepared magnetite nanoparticles (thFs and Fs) was analyzed by a Bruker D2 Phaser X-ray diffractometer (XRD) using Cu-K $\alpha$  radiation operating at 30

**21 kV and 10 mA. Fourier-transform infrared spectroscopy (FTIR) analysis was performed on a**

Shimadzu IRTracer-100. The oxidation states of Fe element in the samples

**1 were determined using an X-ray photoelectron spectroscopy (XPS, Thermo Fischer Scientific, VG ESCALAB 250**

). The magnetic properties were measured by a superconducting quantum interference device (SQUID, MPMS 3) at 300 K. Surface area and pore properties were determined by N<sub>2</sub> adsorption-desorption isotherm procedure using a BEL Belsorp Max Table 1 Synthesis temperature used and its influence on yield and Pi removal ability of the synthesized materials. Data are means  $\pm$  SD from three replicates. Sample name Temp (K) pH\* YieldFe\*\* (%) YieldFe+thiamine\*\*\* (%) % Removal thF-303 303 thF-333 333 thF-363

363 F-303 303 F-333 333 F-363 363 9.24 31.00 § 1.39 ac 9.14 30.04 § 0.55 ab 9.11 33.93 § 1.02 a 9.98  
 29.44 § 0.55 bc 9.92 27.16 § 0.51 bd 9.83 25.72 § 0.30 d 14.00 § 0.64 13.57 § 0.25 15.01 § 0.46 17.09 §  
 1.76 34.37 § 0.88 48.33 § 1.06 3.92 § 0.49 5.82 § 0.46 7.67 § 0.18 \*

**13pH of solution at the end of the reaction. \*\* The amount of product formed per amount of FeSO<sub>4</sub>·7H<sub>2</sub>O reactant used, calculated as Yield<sub>Fe</sub> =  $\frac{m_{\text{product}}}{m_{\text{FeSO}_4 \cdot 7\text{H}_2\text{O}}}$**

\*\*\* 4Th2e

**1 amount of product formed per amount of FeSO<sub>4</sub>·7H<sub>2</sub>O and thiamine reactants used, calculated as Yield<sub>Fe+thiamine</sub>**

$\frac{1}{4}$  mFeSO<sub>4</sub> · 7H<sub>2</sub>O + thiamine 100%. analyzer. Prior to the sorption experimentation, the samples were degassed at 393 K for 4 h. The N<sub>2</sub> adsorption-desorption data (V<sub>a</sub> vs p/p<sub>0</sub>) were then fitted with Brunauer-Emmett-Teller (BET) model, with mathematical expression shown in equation (1).  $\frac{p}{p_0} = \frac{V_a}{V_m} \frac{V_m - V_a}{V_m - V_a p/p_0}$  where V<sub>m</sub> (cm<sup>3</sup>(STP)/g) and V<sub>a</sub> (cm<sup>3</sup>(STP)/g) are the gas volume at the monolayer coverage and the

**26 total gas volume adsorbed at the standard state (T= 273.15 K and P= 101.3 kPa**

), c

**8 is the BET constant and p/p<sub>0</sub> (kPa) is the relative pressure**

. The obtained V<sub>m</sub> value was then utilized to calculate the BET surface area (a<sub>s,BET</sub>, cm<sup>2</sup>/g) by the following equation (2).  $a_{s,BET} = \frac{V_m N_A A_{cs}}{V_m}$  where N is the Avogadro's number (6.023 × 10<sup>23</sup> mol<sup>-1</sup>), A<sub>cs</sub> (cm<sup>2</sup>) is adsorbate cross-sectional area (16.2 × 10<sup>20</sup>), 22414 cm<sup>3</sup>(STP)/mol is the molar volume of gas at STP. The total pore volume (V<sub>p</sub>, cm<sup>3</sup>/g) and the mean pore diameter (d<sub>p</sub>, nm) was determined by equations (3) and (4), respectively.  $V_p = \frac{V_{p,4} - V_{p,1}}{4 - 1} \frac{1000}{a_{s,BET}}$  where V (cm<sup>3</sup>(STP)/g) is amount N<sub>2</sub> adsorbed at last adsorption point, ρ<sub>a</sub> is the density of adsorptive (0.808 g/cm<sup>3</sup>), and M is the nitrogen molecular weight (28.0134 g/mol). The

**1 point of zero charge (pHPZC) determination was done according to the reported procedure**

[35]. In brief, 10 mg of adsorbent

**1 was added into a series of 10 mL**

KNO<sub>3</sub> 0.1 M solution with initial pH between 2 to 10. The initial pH of these KNO<sub>3</sub> solutions was adjusted by HCl 0.1 M or NaOH 0.1 M. Subsequently, the mixture solution was placed on a shaking incubator (200 rpm)

at 303 K. After 36 h incubation, the supernatants were collected and were used for final pH determination. The DpH was calculated by subtracting the value of the final pH from the initial pH. The pHPZC of each particle is the pH where DpH equal to 0. 2.4. Adsorption Study A 10,000 mg/L Pi stock solution

**11 was prepared by dissolving 10 g of KH<sub>2</sub>PO<sub>4</sub> in 1 L of DI water. The**

diluted Pi solution was then prepared from the stock solution and was used for the adsorption experiment. The residual concentration of Pi solution post-adsorption was measured by a colorimetric procedure using a UV-Vis spectrophotometer Shimadzu UV 2600. The Pi solution was filtered using a 0.22 µm PVDF syringe filter membrane prior to the colorimetric measurement. To these filtered solutions, ammonium molybdate-based reagent was added to produce a blue-colored solution which absorbance was measured at 880 nm wavelength [36]. 2.4.1. Screening of Pi adsorption potential Adsorbent (10 mg) was added into 10 mL Pi

**1 solutions at an initial concentration of 100 mg/L, the**

adsorption process lasted for 24 h under constant shaking at 200 rpm, and a temperature of 303 K.

**1 After 24 h, the adsorbent** was magnetically **separated from the**

supernatant, and the concentration of Pi that remained in the supernatant was measured.

**12.4.2. Effect of pH on Pi adsorption The effect of pH on Pi removal was investigated between pH**

range of 2 to 10. NaOH (0.1 M) or HCl (0.1 M) was used to adjust the initial pH of the Pi solution (C<sub>0</sub> = 100 mg/L). Into the 10 mL Pi solution, 10 mg adsorbent was added, and the mixtures was then placed

**2 in a shaking incubator at 200 rpm and**

303 K. After 24 h, the supernatant was separated, and the amount of residual Pi that remained in the supernatant was measured. 2.4.3. Adsorption isotherm The adsorption of Pi by the samples was conducted at pH 3 and 4 for F-363 and thF-363 samples, respectively. A series of 10 mL Pi solutions at a varied concentration (50 to 10,000 mg/L) were prepared in scintillation vials, then 10 mg of the sample was added. These vials were then placed

**10 in a shaking incubator**, which operated **at 200 rpm under a** controlled temperature **of 303**

, 318, or 333K. After 24h, residual Pi in the supernatant was measured. 2.4.4. Data processing The percent removal of Pi (% Removal) and the equilibrium

**24 amount of Pi adsorbed per unit mass of adsorbent (Q<sub>e</sub>)**

where  $Q_e$  (mg/g) and  $C_e$  (mg/L) are the equilibrium concentration of adsorbate in the solution and the equilibrium concentration of adsorbate in the adsorbent, respectively.  $Q_0$  (mg/g) is the maximum adsorption capacity of the adsorbent,  $K_L$  (L/mg) is the Langmuir adsorption equilibrium constant,  $K_F$  (mg/g)(L/mg) $^{1/n}$  is the Freundlich adsorption constant, and  $n$  is the Freundlich adsorption intensity.

where  $C_0$  (mg/L) and  $C_e$  (mg/L) are the initial and equilibrium concentration of  $P_i$ ,  $V$  (L) is the volume of the solution, and  $m$  (g) is the mass of adsorbents. The

adsorption data were plotted as  $Q_e$  vs  $C_e$  and were fitted

against the Langmuir and Freundlich model, which is mathematically expressed as equations (7) and (8), respectively. These two

models are used to respectively describe the monolayer and multilayer adsorption process

that might occur during the adsorbate-adsorbent interaction [37].  $Q_e$  (mg/g) is the equilibrium adsorption capacity,  $K_L$  (L/mg) is the Langmuir adsorption equilibrium constant,  $K_F$  (mg/g)(L/mg) $^{1/n}$  is the Freundlich adsorption constant, and  $n$  is the Freundlich adsorption intensity.

12mg/g) is the Langmuir maximum adsorption capacity,  $K_L$  (L/mg) is the Langmuir adsorption equilibrium constant,  $K_F$  (mg/g)(L/mg) $^{1/n}$  is Freundlich isotherm constant

10related to adsorption capacity, and  $n_F$  is a dimensionless constant related to the favorability of adsorption

. Furthermore, the suitability of adsorbents for  $P_i$  adsorption was evaluated from the value of the constant separation factor ( $R_L$ ) obtained from the following equation (9).  $R_L$  is the separation factor,  $K_L$  (L/mg) is the Langmuir adsorption equilibrium constant, and  $C_0$  (mg/L) is the initial concentration of adsorbate in the solution. Besides the model mentioned above, the Sips three-parameters model was employed for the data fitting purpose. This

2model is a modified form of the Freundlich equation, which follows the continuous increase of capacity as an increase of concentration but has a finite limit at the sufficiently high concentration [38]. The

mathematical expression of the Sips model is given in equation (10).  $Q_e$  (mg/g) is the equilibrium adsorption capacity,  $K_s$  (L/mg) is the Sips adsorption equilibrium constant, and  $n_s$  is the Sips model exponent that expresses the heterogeneity of the adsorbent.

2QS is the Sips maximum adsorption capacity (mg/g),  $K_s$  is the Sips equilibrium constant related to the adsorption affinity (L/mg), and  $n_s$  is the Sips model exponent that expresses the heterogeneity of the adsorbent.

**Sips model reduces to Langmuir model as the  $n=1$  and reduces to Freundlich when either  $C_e$  or  $K_s \rightarrow 0$**

. These isotherm models

**were fitted with the experimental data** gathered to evaluate **the adsorption**

mechanisms to better understand how adsorbates interact with adsorbents. The linear regression (LR), nonlinear regression (NLR), and orthogonal distance regression (ODR) analyses [39, 40] were performed by

**4 minimizing the sum of the squares of the residuals/error** (SSE, equation 11) **while the model coefficients** were **iteratively modified. The**

LR and NLR analyses were

**4 carried out using Microsoft Excel equipped with Solver data analysis tool pack with the regression results assessed through the resulting coefficient of determination ( $R^2$ , equation 12), adjusted R-squared**

(Adjusted, equation 13), while the ODR analysis was conducted with the Origin2019 software.  $X_n$  SSE  $\frac{1}{n} \sum_{i=1}^n (Q_{e,i} - Q_{e,i}^{\text{fit}})^2$   $R^2 = 1 - \frac{\sum_{i=1}^n (Q_{e,i} - Q_{e,i}^{\text{fit}})^2}{\sum_{i=1}^n (Q_{e,i} - \bar{Q}_e)^2}$   $\text{Adjusted } R^2 = 1 - \frac{\sum_{i=1}^n (Q_{e,i} - Q_{e,i}^{\text{fit}})^2}{\sum_{i=1}^n (Q_{e,i} - \bar{Q}_e)^2} \frac{n-1}{n-k}$  where  $Q_{e,i}$  is the fitted value of equilibrium amount of  $P_i$  adsorbed per unit mass of adsorbent (mg/g),  $\bar{Q}_e$  is the average mean value of the equilibrium amount of  $P_i$  adsorbed per unit mass of adsorbent (mg/g),

**$n$  is the number of experimental data points, and  $k$  is the number of parameters in the model, including the equilibrium concentrations predicted. The thermodynamic parameters, i.e. the**

change

**15 in Gibbs free energy ( $\Delta G^\circ$ ), enthalpy ( $\Delta H^\circ$ ), and entropy ( $\Delta S^\circ$ ), were based on the equations (14) to**

(17) [41].  $\Delta G^\circ = -RT \ln K_d$   $\Delta G^\circ = \Delta H^\circ - T\Delta S^\circ$   $\ln K_d = \frac{\Delta S^\circ}{R} - \frac{\Delta H^\circ}{RT}$  where  $K_d$  is the thermodynamic equilibrium constants dimensionless of the adsorption process [42].

**$R$  is the gas constant (8.314 J/mol.K) while  $T$  is the absolute temperature (K). By plotting  $\ln K_d$  versus  $1/T$ , the**

change in  $\Delta H^\circ$ ,  $\Delta S^\circ$ , and  $\Delta G^\circ$  can be obtained. 2.5.

**6Arabidopsis thaliana growth media supplementation** To assess the **environmental** compatibility of

the F-363 and thF- 363, the freshly synthesized materials were administered to the plant growth media [43].

**3A. thaliana Col-0 ecotype was used as the model plant. The**

**6half-strength Murashige and Skoog (MS) basal medium supplemented with 1% (w/v) sucrose, 0.8% (w/v) agar**

media was prepared according to previous publications [3,44-46]. For the F-363 or thF-363 treatment group, 0.1 wt.% of respective materials were added into the basal

**3media. The A. thaliana seeds were germinated and grown**

under

**3continuous light condition at 295 K. After 14 days**

of vertical growth, the aerial fresh weight and

**3primary root length of the seedlings**

were measured. Three independent biological experiments with 8 seedlings per replicate were performed for control, F- 363, and thF-363-treated samples.

**6Statistical analysis was conducted using the GraphPad Prism 8.0 software, and significant differences between**

different growth media compositions

**6were examined by one-way analysis of variance (ANOVA) with Tukey's posthoc test. 3. Results and Discussion**

This study employed three synthesis temperatures (303, 333, and 363 K) for thFs and Fs preparation. To ensure that the suitability of these conditions, firstly, the thiamine thermal stability was tested. As shown in Figure S1, a negligible amount of % mass loss ( $\gg 0.19\%$ ) of thiamine was observed upon 2 h isothermal heating at 363 K (the highest temperature used for the synthesis), which implies that the thiamine remained stable and did not undergo thermal degradation during the synthesis process. The formation of thFs appears

to depend on temperature, as implied from the increase in yield as the reaction temperature was increased (Table 1). The

**20% increase of synthesis temperature from 303 K to 333 K resulted in the**

decrease of thF Yield<sub>Fe</sub> from 31.00% to 30.04%. However, the Yield<sub>Fe</sub> was increased from 30.04% to 33.93% as the synthesis temperature increased from 333 K to 363 K. The increase in synthesis temperature shows a favorable effect on the formation of thF, which could be attributed to the high energy supply at higher temperature, thus facilitating the formation of thF. The supply of energy was reported to affect the nucleation rate in the formation of metal-organic complexes [47,48], which could also be the case in the formation of thF involving the interaction between thiamine and Fe<sub>3</sub>O<sub>4</sub>. Meanwhile, the higher thF-303 Yield<sub>Fe</sub> (compared to Yield<sub>Fe</sub> of thF-333) was attributed to the generation of side product FeO(OH); as shown by the occurrence of two additional XRD peaks at 2θ of 20.81° and 40.21° (Figure S2a), which were attributed to the reported (220) and (420) crystal plane of FeO(OH) (JCPDS No. 01-077-0247) [49,50]. Interestingly, no contaminant peaks were observed in thF-333 and thF-363 samples (Figures S2b and 1a). Besides yield, the synthesis temperature was shown to positively affect the Pi removal efficiency of the resultant thFs, and the highest % Pi removal was achieved upon the usage of thF-363 as the adsorbent. This enhanced adsorption potential might be attributed to the higher composition of thiamine in thF prepared at 363K than the one synthesized at 333K. The TGA plots (Figure S2d and e) confirmed that the thF-363 contained approximately 15.16% thiamine, while only 6.94% of the thF-333 consisted of thiamine. The higher thiamine content in thF-363 provides more adsorption sites for better Pi removal. The proposed interaction between the thF-363 and Pi is described in subsection 3.3. Based on the above-mentioned results, thF-363 has better Pi removal efficiency and higher Yield<sub>Fe</sub> than thF-303 and thF-333; thus, detailed characterization and Pi adsorption studies were carried out for thF-363 and F-363 (as the control).

### 3.1. Characterization of thF-363

Figure 1a shows the crystal pattern of F-363 and thF-363; both samples had seven distinct diffraction peaks at similar 2θ angles, and these XRD patterns match well with the XRD pattern of Fe<sub>3</sub>O<sub>4</sub> reference (JCPDS 19-0629), indicating the successful formation of Fe<sub>3</sub>O<sub>4</sub> particles. The crystallinity of thF-363 was 57.8%, which is significantly lesser than the F-363 (81.2%). This reduced crystallinity can be attributed to the incorporation of thiamine on the magnetite core. The formation of Fe oxide was also confirmed by the occurrence of Fe-O functional

**20 group at 576 cm<sup>-1</sup> and 573 cm<sup>-1</sup> in the**

thF-363 and F-363 FTIR spectra (Figure 1b). Beside the Fe peak, thiamine fingerprint spectra, specifically the N-H group (3314

**25 cm<sup>-1</sup>), C-H aliphatic group (2922 cm<sup>-1</sup>), aromatic amine C**

-N group (1652 cm<sup>-1</sup>), C=C group (1524 cm<sup>-1</sup>), C-S group (1325 cm<sup>-1</sup>), and C-O group (1037 cm<sup>-1</sup>)

**2 can be observed in the FTIR spectra of**

thF-363 which verify the formation of thiamine-functionalized Fe<sub>3</sub>O<sub>4</sub>. XPS analysis was performed to investigate the elemental state of Fe ions in thF-363 and F-363. As shown in Figure 1c, three peaks of iron (Fe 2p) with binding energies at 710.8, 712.3, and 724.8 eV, respectively attributed to Fe<sup>3+</sup> 2p<sub>3/2</sub>, Fe<sup>2+</sup> 2p<sub>3/2</sub> and Fe 2p<sub>1/2</sub>, can be observed in the XPS patterns of F-363 [51,52]. Similarly, the thF-363 samples also showed three Fe 2p-related XPS spectra with the binding energy of 710.7, 712.6, and 724.7 eV (Figure

1d). Based on the stoichiometric, the Fe<sup>2+</sup>:Fe<sup>3+</sup> ratio of thF-363 and F-363 is shown to be 0.34:0.66 and 0.37:0.63, respectively. These ratios are comparable to the previously reported Fe<sub>3</sub>O<sub>4</sub> Figure 1. (a) Powder XRD pattern and (b) FTIR spectra of the thiamine-functionalized Fe<sub>3</sub>O<sub>4</sub> synthesized at 363 K (thF-363) and control; unmodified Fe<sub>3</sub>O<sub>4</sub> synthesized at 363 K (F-363). (c,d) The XPS spectra of (c) F-363 and (d) thF-363. (e) The N<sub>2</sub> adsorption-desorption isotherms curves and (f) hysteresis curves showing the magnetic properties of F-363 (black) and thF-363 (blue). data [52], suggesting that the iron oxide in thF-363 and F-363 are in Fe<sub>3</sub>O<sub>4</sub> form. The N<sub>2</sub> adsorption-desorption isotherms of thF-363 and F-363 are shown in Figure 1e. According to IUPAC classification [53], the isotherm peak of both samples exhibited the type IV isotherms curve with H3-types hysteresis loop, which is commonly observed for mesoporous materials. Upon fitting by BET model, the dp of thF-363 and F-363 is 32.913 nm and 36.976 nm, respectively, which is within the range of mesoporous material (2 to 50 nm). In addition, the thF-363 particles have an as,BET of 32.999 m<sup>2</sup>/g that is greater than that of the control (27.490 m<sup>2</sup>/g). Figure 1f displays the magnetic hysteresis curves of thF-363 and F-363 measured at 300 K. The saturation magnetization values (M<sub>s</sub>) of thF-363 and F-363 is 84.67 and 91.71 emu/g. Lower M<sub>s</sub> value of thF-363 might be due to the addition of thiamine which is non-magnetic. Despite the smaller M<sub>s</sub> value, thF-363 (and F-363) fell into the classification of superparamagnetic material. Disregard of the reduced M<sub>s</sub>,

**1 these values are postulated to be sufficient for magnetic recovery from solution**

[54]. 3.2. Effect of initial pH on Pi adsorption The influence of initial pH on the Pi removal by thF-363 and F-363 adsorbent was presented in Figures 2a and 2b. Optimum Pi removal by using thF-363 can be achieved when the initial pH of the adsorbate was set at pH 4.

**3 On the other hand, the efficiency of Pi removal by**

F-363 decreased upon increasing pH, and the optimum removal can be obtained between pH 2-3. The pH for Pi adsorption is essentially concentrated at low pH for both samples, which can be Figure 2. Effect of solution pH on Pi adsorption onto (a) thF-363 and (b) F-363. (

**1c) The pH of point of zero charge of thF-363 and**

F-363. Data are means ± SD from three replicates.

**1 Different lowercase letters above the graph bars denote significant differences among different pH (ANOVA and Tukey's test, p<0.05).** explained by observing **the pHPZC of**

thF-363 and F-363 adsorbent. As shown in Figure 2c, the pHPZC value for thF-363 and F-363 is 5.6 and 4.4, respectively. The pHPZC value indicates the surface charge tendency of the adsorbent; positive surface charge occurs at pH solution < pHPZC, while negative surface charge occurs at pH solution > pHPZC [55]. The interaction of Pi and thF-363 at acidic and basic pH is illustrated in Figure 3. At acidic pH (i.e., pH solution < pHPZC), thF-363 particles are positively charged while Pi dominantly presents as negatively charged species (H<sub>2</sub>PO<sub>4</sub><sup>-</sup>) [56]. The opposite charges between the adsorbent and adsorbate generate electrostatic attraction, which promotes the adsorption of Pi onto the adsorbent surface. At basic pH (i.e., pH solution > pHPZC), thF-363 tends to be negatively charged; and, Pi dominantly occurs as negatively

charged species of  $\text{HPO}_4^{2-}$  [56]. The same charges between thF-363 and Pi lead to repulsion force, which demotes the adsorption of Pi. A similar effect of surface charge on the adsorption performance of adsorbent has also been reported in other works [46,57,58]. For example, Jiang et al. [57] Figure 3. Illustration on the effect of pH, adsorbent surface charge, and adsorbate species distribution on the thF-363 and Pi interaction. Figure 4. Isotherm data of Pi adsorption on (a,c,e) thF-363 and (b,d,f) F-363 at three different adsorption temperatures (303 K, green; 318 K, blue; and 333 K, red symbols). The experiments were done in triplicate. The lines represent the (a, b) Langmuir, (c, d) Freundlich, and (e,f) Sips data fitting. reported that the  $\text{Fe}_3\text{O}_4$  particles were

10 **negatively charged at pH 7**, which led to **the** electrostatic **adsorption** between **the**

particles and positively charged anthocyanin. In addition, the occurrence of excess hydroxyl ions also may act as competing molecules and occupy the adsorption site of Pi anions on the adsorbent [59]. 3.3. Adsorption isotherm of Pi onto thF-363 magnetic adsorbent The adsorption isotherm

21 **plays a crucial role in** determining **the** maximum **adsorption capacity and**

estimating the adsorption mechanism of adsorbents. The isotherm study of Pi on thF-363 and F-363 was conducted at adsorption temperatures of 303, 318, and 333 K (Figure 4). At any given temperature, the system containing thF-363 displayed typical H-adsorption curves, which suggests that the adsorption was driven by the electrostatic forces due to different charges of adsorbate and adsorbent. While for F-363 adsorbent, the system shows an L-type curve which commonly occurs in the adsorption of ions in aqueous solutions and

1 **the adsorption process** is **driven by van der Waals forces**

[60]. Overall, the adsorption process with an adsorbent plateau at high adsorbate concentration follows the sub-class classification number 2, demonstrating the absence of the inter-molecular forces

2 **between the adsorbed-adsorbate molecules and the adsorbate molecules in bulk solution**

[61]. Both thF-363 and F-363 tend to exhibit endothermic adsorption behavior, where the equilibrium adsorption capacity ( $Q_e$ ) increases with temperature. Three isotherm models, Langmuir, Freundlich, and Sips, were used to evaluate the adsorption behavior of Pi onto magnetite sorbent thF-363 and F-363; the calculated parameters of NLR fitting the isotherm models are listed in Table 2. The NLR fitting method was chosen as the best regression method since it gave the smallest SSE value and the closest fitting to the experimental data compared to LR and ODR methods (Tables S1, S2, and Figures S3, S4). The Sips model showed the best fitting with the experimental data of both thF-363 ( $\text{Rad}j^2 = 0.988\text{--}0.994$ ) and F-363 ( $\text{Rad}j^2 = 0.997\text{--}0.999$ ). The goodness of fitting also is displayed on the value of calculated QS from the Sips model, which showed the closest resemblance to the  $Q_{\text{exp}}$ . The Sips Table 2 Adsorption isotherm parameters of Pi adsorption on either thF-363 or F-363 obtained from nonlinear regression fitting using Langmuir, Freundlich, and Sips models. Model Parameter (Unit) thF-363 F-363

22303 K 318 K 333 K 303 K 318 K 333 K Langmuir Freundlich

Sips  $Q_{exp}$ (mg/g) QL (mg/g) KL (L/mg) RL R<sub>2</sub> Radj 2 SSE KF (mg/g)(L/mg)?n nF R<sub>2</sub> Radj 2 SSE QS (mg/g) KS (L/g) nS R<sub>2</sub> Radj 2 SSE 221.993 226.051 0.0042 0.02-0.83 0.994 0.993 559.782 39.190 5.017 0.938 0.933 5553.256 225.837 0.004 1.005 0.994 0.993 559.683 242.281 243.906 0.0072 0.01-0.74 0.987 0.986 1443.821 57.528 5.866 0.923 0.918 8802.450 257.013 0.025 0.748 0.989 0.988 1211.164 280.954 286.376 0.0086 0.01-0.70 0.994 0.994 911.814 67.881 5.854 0.886 0.879 18520.144 287.680 0.010 0.967 0.994 0.994 905.877 146.977 177.699 0.0005 0.17-0.98 0.997 0.997 139.427 3.519 2.404 0.951 0.948 2232.027 168.019 0.0003 1.108 0.998 0.997 101.864 206.093 248.289 0.0007 0.13-0.97 0.982 0.980 1811.293 8.388 2.743 0.894 0.887 10464.750 209.278 0.00001 1.695 0.999 0.999 70.988 230.923 275.639 0.0009 0.1-0.96 0.983 0.981 2575.329 9.833 2.752 0.875 0.868 17976.803 238.421 0.00002 1.594 0.999 0.999 156.020

model also

**3 can be used to confirm the satisfactory of the Langmuir or Freundlich fitting. Based on the fitting results, the nS value of all the adsorption systems is close to 1, indicating their better approach to the Langmuir model. Furthermore, the KS of the Sips model does not show zero value, which confirms the Langmuir approach**

[35,62]. In agreement with the Sips fitting result, the Langmuir model showed better fitting with the experimental data for both adsorbents, while the fitting with Freundlich model tends to deviate strongly from the experimental data. Based on the Langmuir isotherm, the maximum adsorption capacity of Pi increased from 226.051 to 286.376 mg/g for thF-363 and 177.699 to 275.639 mg/g for F-363 at the increasing adsorption temperature. Proportionally, the equilibrium constant KL value also increased at higher temperature. On the other hand, the favorability of adsorbent towards Pi was evaluated by the value of the separation factor constant (RL) which can be obtained from Langmuir model. The value of RL of all adsorption experiments showed the value between 0 and 1, indicating that the synthesized adsorbents were all suitable for the Pi adsorption [63]. Disregards of the data fitting, the nF values of both samples fallen within the 2-10 range which indicate the adsorption is favorable [63]. It is also worth mentioning that the thF-363 showed substantially higher adsorption capacity compared to F-363; up to 1.51-fold higher capacity for adsorption at 303K. The mechanism of Pi adsorption on F-363 and thF-363 is illustrated in Figure 5. The adsorption of Pi on F-363 was driven by the electrostatic attraction between the anionic species of Pi and positively charged F-363, which is a typical phenomenon observed in the adsorption of anionic Pi onto iron oxides-based sorbents [64,65]. Distinctively, the enhanced adsorption capacity in thF-363 adsorbent may be attributed to the presence of thiamine, which provides additional amino ( $\text{NH}_2$ ) binding sites. The high Pi adsorption on the  $\text{NH}_2$  functionalized adsorbents has been reported in several works [66-68], wherein the anionic Pi establishes electrostatic attraction with the protonated species of the  $\text{NH}_2$  functional group. Pi removal through adsorption method had been previously studied using various magnetite-based adsorbents [66,68-71]. A comparison of the maximum Pi adsorption capacities onto thF-363, F-363, and other magnetite composite adsorbents is presented in Table 3. The capacity of thF-363 for Pi adsorption is significantly higher than other reported magnetite adsorbents, which show the superiority of thF-363 adsorbent.

### 3.4. Adsorption thermodynamic

Thermodynamic parameters can be used to estimate the characteristics of Pi adsorption onto the adsorbent surface. Figure 6 shows the adsorption thermodynamic of the adsorbents. The negative  $\Delta G^\circ$  Figure 5.

**3 Possible mechanism of Pi adsorption on (a) F-363 and (b**

) thF-363. Table 3 Comparison of Pi adsorption capacities and optimum adsorption condition of thF- 363, F-363, and previously reported magnetite-based adsorbents. Adsorbent Conditions Qmax (mg/g) References  
 thF-363 F-363 Fe3O4/ZrO2/chitosan Fe3O4@LDH composites Fe3O4@alkali-treated calcium-silicate  
 Chitosan/Al2O3/Fe3O4 nanofiber Rectorite/Fe3O4-CTAB pH 4, 333 K pH 3, 333 K pH 3, 298 K pH 3 pH 8,  
 298 K pH 3, 303 K pH 5, 303 K 287.7 238.4 26.5 26.5?36.9 128 130.9 174.5 This study This study [68] [69]  
 [70] [66] [71] value at all given temperatures demonstrate the thermodynamically favorable and spontaneous  
 adsorption of Pi onto the thF-363 and F- 363 surface. Specifically -26.22, -28.47, and -30.72 kJ/mol for the  
 DGo of thF-363 at 303, 318, and 333 K, and DG -20.83, -22.67, and -24.52 kJ/mol for F-363 at 303, 318,  
 and 333 K. The DH for each tem- perature increase was evaluated to observe the heat transfer direction in  
 the adsorption system. Positive DH values obtained in the Pi?thF-363 system was 27.66 kJ/mol, 10.24

**23kJ/mol and 19.23 kJ/mol** when **the adsorption** temperature **was** increased from  
 303 **to**

318K, 318 to 333 K, and 303 to 333 K, respectively. The same positive values of DH were cal-  
 culated from the system containing F-363 as well (DH = 18.84 kJ/mol for 303 ! 318 K, 13.89 kJ/mol for 318 ! 333 K, and  
 16.44 kJ/mol for 303 ! 333 K). These positive DH values indicate that the adsorption pro-  
 cess happens endothermically and favors high temperature [58,72], as confirmed by the maximum adsorption capacity  
 increase when the tem- perature increases from 303 to 333 K (Table 2). The DS value for each temperature  
 increase was also calculated. A positive value of DS was found at each temperature increase. For instance,  
 the DS = 0.178 kJ/mol K and 0.150 kJ/mol K when the adsorption temperature in Pi?thF-363 system  
 increased from 303 to 318K and 303 to 333K, respectively. In the system containing F-363, the S303 318  
 was 0.131 kJ/mol K and S303 333 is 0.123 kJ/mol K. D ! D ! The

**15positive DS values indicate** the **increase in the** disorder **of** ions in **the**  
 adsorbate-adsorbent **interface**

along with the increase in temper- ature [63,73]. 3.5. Environmental compatibility assessment of thF-363 on  
 plant growth The usage of non-environmentally friendly adsorbents for con- taminant removal from  
 wastewater may evoke ecological risk toward soils, plants, and their surrounding ecosystem. To evaluate the  
 envi- ronmental compatibility of thF-363, this work focused on its effect on the overall growth of *A. thaliana*  
 seedlings upon supplementation to plant growth media. The evaluation was based on phenotypical  
 observation of the seedling aerial and root development. Figure 7a Figure 6. Adsorption thermodynamics for  
 the removal of Pi by (a-c) thF-363 and (d-f) F-363. Figure 7.

**3Growth observation of Arabidopsis thaliana seedlings grown on**  
 supplemented **media. (a)** Image **of** 14-day-**old seedlings. (b)**

) Aerial fresh weight (upper panel) and

**3primary root length** (lower panel) **of the seedlings**

. Data are means § SD from three replicates, with 8 seedlings observed per replicate. Different letters above  
 and below the

**1** graph bars denote significant differences among different growth media (ANOVA and Tukey's test,  $p < 0.05$ ). shows the

representative 14-day-old seedlings grown on basal (control), thF-363 supplemented (thF), and F-363 supplemented (F) media. The measured aerial fresh weight and primary root length of the seedlings are presented in Figure 7b. No observable difference can be seen between the aerial fresh weight of seedlings grown on the thF, F, and control. However, the thF-363 supplementation slightly affects the primary root growth of the seedlings (Figure 7b, lower panel). This phenomenon might be due to the high adsorption capacity of freshly synthesized thF-363, which reduces nutrient availability in the media, thus inhibiting root growth. Nevertheless, both modified and unmodified Fe<sub>3</sub>O<sub>4</sub> caused minimal to no adverse effects on phenotypical growth of seedlings' root and aerial tissues, which suggest their non-phytotoxic properties. mechanisms. Both modified and unmodified Fe<sub>3</sub>O<sub>4</sub> showed a non-toxic effect on plant growth, confirming their environmentally-friendly nature. Author Contribution

**1A.E.A. Conceptualization; Data curation; Funding acquisition; Investigation; Writing.** Y.N.T.-C. **Formal analysis; Investigation;** Methodology; **Visualization; Writing.** Q.N.H. **Formal analysis;** Investigation; **Methodology**

. P.L.T.-N. Funding acquisition; Supervision; Project administration. S.P.S., and

**1A.W.G. Supervision; Project administration; Resources. V.B**

. Formal analysis. H.-Y. H and Y.-H.J. Supervision; Resources. 4. Conclusion An environmentally friendly adsorbent with high adsorption capacity toward phosphate ions (Pi), namely thiamine-functionalized Fe<sub>3</sub>O<sub>4</sub> (thF), was successfully synthesized through in-situ reaction by combining NH<sub>4</sub>OH:Fe:thiamine at a molar ratio of 10:1:1 and a temperature of 363 K. Detailed characterization through XRD, XPS, FTIR, SQUID, and N<sub>2</sub> adsorption-desorption isotherm confirmed the formation of Fe<sub>3</sub>O<sub>4</sub>, the incorporation of thiamine, and gave insight on the other physicochemical properties of the thF-363. The thiamine functionalization was shown to substantially increase the adsorption capacity for Pi removal up to 1.51-fold (at 303 K) compared to the unmodified Fe<sub>3</sub>O<sub>4</sub>. The improved adsorption capacity is attributed to the synergistic effect of the surface charge, area, and the occurrence of the thiamine functional group in thF-363. The

**27** adsorption process fitted well with the Langmuir and Sips isotherm model, with a maximum adsorption capacity of 287.680 mg/g at 333 K. The

Pi adsorption to the adsorbents followed endothermic and spontaneous Declaration of Competing Interest The authors declare that the research was conducted without

**28** any commercial or financial relationships that could be construed as a potential conflict of interest. Acknowledgments

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