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RESEARCH ARTICLE

Utilization of iron scrap into copperas and rambutan (Nephelium lappaceum L.) peel extract into environmentally friendly iron nanoparticles for hexavalent chromium removal and its kinetics

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Abstract

Hexavalent chromium is a toxic pollutant that is very harmful to human health and the environment. One of the materials for hexavalent chromium removal is iron nanoparticles (INPs). In recent years, the manufacture of environmentally friendly INPs has been of great interest. In general, INPs are made using sodium tetra borane/NaBH $_4$, which is expensive. The synthesis of INPs using N aBH₄ also produces side products in the form of toxic boric acid and hydrogen gas which are explosive and flammable. In this study, INPs are made by using iron scrap waste into copperas which is then reacted with rambutan peel extract (RPE). The reaction between iron scrap and sulfuric acid until crystals are formed and then washed until blue-green crystals are formed. The resulting crystal is made into a solution and then reacted with RPE. After forming a black solution, it was dried by spray drying. The resulting black powder was characterized by UV-Vis Spectrophotometer, SEM-EDX, FTIR, and XRD. The results showed that iron scrap can be synthesized into copperas with a Fe content of 19.84 ± 0.15 %. RPE has a total phenol content of 877.39 ± 16.6 ppm/100 g rambutan skin or equivalent to 441.42 mg GAE/100 g rambutan skin. INPs can be made from copperas from iron scrap and RPE which have Surface Plasmon Resonance (SPR) characteristics at 214 nm absorption, particle size 20–70 nm. Hexavalent chromium removal using INPs was maximum at pH 3, hexavalent chromium concentration of 5 ppm, INPs concentration of 0.15 g/100 mL, and contact time of 10 min. The adsorption isotherm type of hexavalent chromium using iron nanoparticles can take place chemically or physically. The correlation coefficient showed that the model of Freundlich (R² > 1.99) is more suitable than the model of Langmuir ($R^2 = 0.98$) with an adsorption capacity of 91.74 mg iron nanoparticles/g hexavalent chromium. The use of INPs for hexavalent chromium removal is a promising and effective technology. These results can be an alternative to the production of environmentally friendly INPs by using materials from waste into useful materials for the prevention of environmental pollution.

KEYWORDS

environmentally friendly, iron nanoparticles, iron scrap, rambutan peel extract

1 INTRODUCTION

In recent years, research on iron nanoparticles (INPs) has received great attention. This is because nanoparticles are an important material to overcome environmental pollution. The use of INPs is very broad including for the treatment of water, wastewater, wells, soil, sediment, and gas flow (Machado et al., [2013\)](#page-11-0). The. For instance, ciprofloxacin, sulfamethoxazole technology of using INPs provides potential advantages over conventional methods due to their unique physicochemical properties, non-toxicity, and economics (Etemadi et al., [2017;](#page-10-0) Fazlzadeh et al., [2017;](#page-10-0) Poguberović et al., [2016;](#page-11-0) Ravikumar et al., [2016\)](#page-11-0). The main advantage of this technique is that relatively simple using only two reagents and no need for specialized equipment. In addition, since it is very small, it can be effectively injected and transported into groundwater and polluted aquifers in situ treatment (Chang et al., [2014;](#page-10-0) Chen et al., [2015;](#page-10-0) Li et al., [2017;](#page-11-0) Madhavi et al., [2013\)](#page-11-0). The pollutant that can be remediated by INPs include azo compounds, chlorinated pesticides, chlorinated solvents, transition metals, and inorganic anions (Crane & Scott, [2012\)](#page-10-0), inorganic and organic pollutants (Shahwan et al., [2011\)](#page-11-0), heavy metals (Li et al., [2017;](#page-11-0) Yuan et al., [2016\)](#page-12-0), nitrate (Wang et al., [2014\)](#page-12-0), bromide (Xu et al., [2015\)](#page-12-0), arsenic (Poguberović et al., [2016;](#page-11-0) Prasad et al., [2014\)](#page-11-0), hexavalent chromium (Chen et al., [2015;](#page-10-0) Jiao et al., [2015;](#page-11-0) Poguberović et al., [2016;](#page-11-0) Ravikumar et al., [2016;](#page-11-0) Sheng et al., [2016\)](#page-11-0), lead (Kumari et al., [2015\)](#page-11-0), antibiotics (Leili et al., [2018\)](#page-11-0), and colorants (Fujioka et al., [2016\)](#page-10-0). INPs are also used in cosmetics, biomedicine, bioremediation, clinical materials, and engineering (Harshiny et al., [2015\)](#page-10-0). INPs have unique properties such as small particle size, and large surface area, making them highly reactive (Mystrioti et al., [2016;](#page-11-0) O'Carroll et al., [2013\)](#page-11-0), catalytic and optical, electronic and antibacterial activities (Harshiny et al., [2015;](#page-10-0) Kokila et al., [2015;](#page-11-0) Saif et al., [2016\)](#page-11-0).

The technology of using INPs provides potential advantages over conventional methods due to their unique physicochemical properties, non-toxicity, and economics

Nanoparticle production is generally categorized into two groups: top-down and bottom-up approaches. The top-down approach is to break down large particles into nanometer-size particles. The bottomup approach is to start with atoms or molecules or clusters that are engineered to form the desired nanometer-size particles (Devatha et al., [2016;](#page-10-0) Saif et al., [2016\)](#page-11-0). Synthesis of INPs with a top-down approach including the preparation of nanoparticles by physical methods, thermal breakdown, sonochemical synthesis, and vacuum sputtering. These methods have limitations, namely, low production rates,

high temperatures and pressures, or high energy requirements, making them relatively expensive (Devatha et al., [2016;](#page-10-0) Mo et al., [2015;](#page-11-0) Wang et al., [2017\)](#page-12-0).

Production of INPs with an approach of a bottom-up including production methods of traditional. Some methods of traditional methods of producing INPs include coprecipitation (Petcharoen & Sirivat, [2012;](#page-11-0) Yoon et al., [2014\)](#page-12-0), hydrothermal method (Giri et al., [2005\)](#page-10-0), and hydrolysis (Tsang et al., [2006\)](#page-12-0). In general, the preparation of INPs is done by reducing iron valence Fe^{3+}/Fe^{2+} with sodium tetra borane (NaBH₄) (Prasad et al., [2014\)](#page-11-0). Preparation of INPs using NaBH4 produces toxic boric acid and hydrogen gas as by-products that are easily explosive and flammable (Ahame $\frac{21}{3}$ al., [2016;](#page-10-0) Devatha et al., 2016; Saif et al., [2016\)](#page-11-0).

Meanwhile, lathe activities produce waste in the form of iron chips. Solid waste from the lathe workshop has been mostly utilized for recycled iron with mark very low economy value. Iron powder from the lathe workshop which is very small in size is usually not utilized anymore, disposed of directly and this causes environmental pollution. The waste still contains valuable materials that when recycled can provide an economic return, namely, with the principles of reuse, recycling, and recovery.

On the other hand, rambutan fruit skin is waste that is simply thrown away. Rambutan peels contain bioactive compounds such as polyphenols (Anoua $\frac{4}{\epsilon}$ l al., [2015;](#page-10-0) Isacfranklin et al., [2020;](#page-11-0) Karnan et al., [2016;](#page-11-0) Karnan & Selvakumar, [2016;](#page-11-0) Mendez-Flores et al., [2018;](#page-11-0) Thitilertdecha & Rakariyatham, [2011;](#page-11-0) Thitilertdecha et al., [2010;](#page-11-0) Yuvakkumar et al., [2015;](#page-12-0) Zhuang et al., [2017\)](#page-12-0). Polyphenols are compounds that can reduce Fe²⁺ to Fe⁰ (Lei<mark>.20,</mark> al., [2018;](#page-11-0) Soliemanzadeh <mark>et al.,</mark> [2016;](#page-11-0) Wei et al., [2016\)](#page-12-0). The amount of rambutan peel is about 25% of whole fruit and is a waste product is a potential material as a source of polyphenols (Karnan & Selvakumar, [2016;](#page-11-0) Kumar et al., [2016;](#page-11-0) Thitilertdecha et al., [2008,](#page-12-0) [2010\)](#page-11-0),which can be used for the manufacture of environmentally friendly INPs. The polyphenol content in RPE is thought to be able to reduce $Fe²⁺$ from copperas to INPs.

The general objective of this research is to remove hexavalent chromium using INPs synthesized from RPE and ferrous sulfate from iron waste. The general objective can be achieved by answering the specific objectives, namely: (1) determining the copperas content of iron scrap, (2) determining the total phenol content of RPE, (3) characterizing INPs made from copperas base of iron scrap and RPE, and (4) reducing hexavalent chromium using INPs synthesized from RPE and ferrous sulfate from iron waste.

2 MATERIALS AND METHODS

The materials used in the study were iron scrap taken from a lathe workshop. The rambutans were bought from the fruit market. Iron nanoparticle samples were obtained by reacting copperas from iron scrap with rambutan peel extract (RPE). The source of chromium was Potassium Chromate (K_2 CrO₄) from Sigma Aldrich. The source of concentrated sulfuric acid for making copperas from Merck. The tools used in research are UV-Vis Spectrophotometer (Shimadzu

UV-1650pc Spectrophotometer),²⁹ canning Electron Microscope-Electron Dispersive X-Ray Spectroscopy (SEM-EDX) brand Shimadzu type 600, Fourier Transform Infra Red (FTIR, Nicolet 6700, USA), and X-Ray Diffraction (XRD) Shimadzu type 600.

The materials used in the study were iron scrap taken from a lathe workshop

2.1 Preparation and determination of copperas content from iron scrap

Preparation of copperas from iron scrap from the lathe workshop as follows: Approximately 25 g of clean iron scrap was reacted with 100 mL of 25% sulfuric acid in an Erlenmeyer. The solution was filtered and then put in the refrigerator for 24 h. The crystals formed were filtered with a vacuum pump and then washed with alcohol. The crystals formed were dried in a desiccator. Determination of levels was carried out by UvVis Spectrophotometer.

2.2 Preparation of rambutan peel extract (RPE) and determination of phenol total content

Preparation of RPE as a source of polyphenols using the method by Kokila et al. [\(2015\)](#page-11-0). The 100 g rambutan peel was cut into small pieces with a size of ± 0.5 cm and boiled in distilled water for 90 min at a temperature of $\pm 800^{\circ}$ C. The ratio of rambutan peel mass to water mass is 1:3. The filtrate was filtered with a filter cloth, then with Whatman paper no. 1 on vacuum filtration. A qualitative test for phenol was done by looking at the presence of the OH group using FTIR at 3200–3550 cm−¹ (Kokila et al., [2015\)](#page-11-0).

2.3 Preparation and characterization of INPs

Preparation of INPs was carried out using copperas from iron scrap and RPE. For the preparation of INPs, the copperas solution used was 5 mL of 0.001 M added to 5 mL of RPE. The reaction was carried out with constant stirring and at room temperature for 5 min (Anouar et al., [2015\)](#page-10-0). The formation of a black solution indicates the formation of INPs (Fazlzadeh et al., [2017;](#page-10-0) Harshiny et al., [2015;](#page-10-0) Karnan & Sel-vakumar, [2016;](#page-11-0) Kuma**r li**dal., [2015;](#page-11-0) Machado et al., [2013;](#page-11-0) Noruzi & Mousivand, [2015;](#page-11-0) Ravikumar et al., [2016;](#page-11-0) Shahwan et al., [2011;](#page-11-0) Wei et al., [2016\)](#page-12-0). The formed colloidal INPs were dried by spray drying. The crystals formed were alracterized by UV-Vis Spectrophotometer, SEM-EDX, FTIR, and XRD.

2.4 Determination of efficiency and adsorption type of hexavalent chromium removal by INPs

In each experiment, 100 mL of hexavalent chromium solution (5, 10, 15) ppm was added (0.01, 0.05, 0.1, 0.15) g INPs, varying pH (3, 7, 11). The mixture was stirred with a stirring speed of 150 rpm at room temperature and time variations (3, 5, 10, 15, 20, and 30) min. The concentration of hexavalent chromium in the filtrate was analyzed using 1,5 diphenylcarbazide an¹⁹ easured by Uv-Vis spectrophotometer at a wavelength of 540 nm. The adsorbed hexavalent chromium was calculated from the initial hexavalent chromium concentration minus the hexavalent chromium concentration after adsorption in the filtrate. The performance of INPs was determined by constructing a curve between the percentage of hexavalent chromium adsorbed and contact time.

The hexavalent chromium removal percentage was calculated by the formula:

% Removal =
$$
\frac{C_i - C_f}{C_i} \times 100\%
$$
 (1)

The optimum results of the performance test of iron nanoparticles that have been obtained are then removed with variations in adsorbate concentration to determine the type of adsorption isotherm. Into five 250 mL beakers, each filled with 0.1 g of iron nanoparticles at optimum conditions, then added hexavalent chromium solution with a concentration of (5, 10, 15, 20, 25) ppm 100 mL as the initial concentration (C0), then stirred at the optimum time. The removal results were filtered, and then the filtrate was measured for hexavalent chromium content with a UV-Vis spectrophotometer (Ca).

 $\frac{8}{2}$ he adsorption capacity (qe, mg/g) was calculated by Equation (2):

$$
qe = \frac{c_0 - c_e}{m} \times V \tag{2}
$$

V(L), volume of solution chromium hexavalent, m (mg) for mass INPs. The ⁵ eundlich and Langmuir isotherms are used to explain the balance between adsorbate and adsorbent using Equations (3) and (4).

$$
\log qe = \log Kf + \frac{1}{n} \log Ce \tag{3}
$$

$$
\frac{Ce}{qe} = \frac{1}{qmax.b} + \frac{Ce}{qmax}
$$
 (4)

caption,

qe, mass adsorbate adsorbed by the asorbent (mg/g)

Kf, factor Freundlich capacity (determined by qe)

Ce, the equilibrium concentration adsorbate in phase liquid after adsorption (mg/L)

n, the Freundlich intensity parameter

qmax, the amount of **maximum** possible adsorbate absorbed (mg/kg)

b is constant Langmuir isotherm.

EXHIBIT 1 Iron scrap and copperas from iron scrap. [Color figure can be viewed at wileyonlinelibrary.com]

EXHIBIT 2 Determination of copperas content of scrap iron.

						Rate determination Λ_{max} (n.m.) absorbance [Fe ²⁺] (ppm) Weight Fe ²⁺ (mg) Fe ²⁺ content (%) Copperas weight (mg) Copperas content (%)	
	510	0.096	0.47	59.10	19.73	294.21	98.23
		0.097	0.48	59.77	19.94	297.53	99.27
Average (x)					19.84		98.75
Standard deviation (s)					0.15		0.74

Then made a graph log *qe* to the log Ce for the Freundlich and Ce/qe isotherms to *Ce* for Langmuir isotherm.

3 RESULTS AND DISCUSSION

3.1 Preparation and determination of copperas content

The reaction between iron scrap from the lathe workshop and 25% sulfuric acid produced blue-green crystals as shown in Exhibit 1.

The reaction for making copperas from iron scrap and sulfuric acid is:

$$
Fe_{(s)} + H_2SO_{4(I)} \rightarrow Fe^{2+}_{(aq)} + SO_4{}^2_{(aq)}^-
$$

$$
H_2SO_{4(I)}\to 2H^+_{(aq)}+SO_4{}^2^-_{(aq)}
$$

$$
Fe_{(s)} + 2H^+_{(aq)} \rightarrow Fe^{2+}_{(aq)} + H_{2(g)}
$$

 $FeSO_{4(aq)} + H_2SO_{4(aq)} + 14H_2O \rightarrow 2FeSO_4.7H_2O_{(s)} + H_{2(g)}$

The determination results from copperas levels from iron scrap was done with the use of spectrophotometry Uv -Vis as shown in Exhibit 2.

Exhibit 2 shows that the average Fe^{2+} content in copperas is 19.84 \pm 0.15% while the average content of copperas from iron scrap

is 98.75 \pm 0.74%. This shows that the copperas synthesized from iron scrap in the lathe workshop meet the quality requirements of copperas SNI 06-4888-1998 which requires a minimum Fe content of 19% (BSN, 1998; Valentovic, 2007).

3.2 Determination of total phenolics of rambutan peel extract

Exhibit [3](#page-5-0) shows the results of the calculation of total polyphenol content of RPE.

Exhibit [3](#page-5-0) shows that the average TPC was 877.40 ± 16.6 ppm/100 g rambutan peel. So the total polyphenol content in RPE is 877.39 ± 16.6 ppm.

The total phenol concentration in the extract was the $\frac{3}{3}$ sed for the calculation of the equivalent organic carbon concentration in the extract, OC (GAE), in mg C/L, by considering the presence of a carbon atom in each gallic acid molecule, according to the following equation:

$$
OC(GAE) = \left[\frac{TPC}{M}\right] \frac{X}{X12g/mol}
$$

description:

TPC: Total Phenolic Content (total polyphenol concentration) (mg GAE/L)

 $\frac{1}{2}$ olecular weight of gallic acid (C₇H₆O₅), equal to 170.12 g/mol 7: Number of carbon atoms in each gallic acid molecule

12 g/mol: Weight of carbon atoms

(Mystrioti et al., [2016\)](#page-11-0). Based on the calculation results, the total phenol content in RPE is 441.43 mg GAE/100 g rambutan skin.

EXHIBIT 3 Total phenol content of RPE.

EXHIBIT 4 Reaction between copperas solution and RPE to produce INPs.²⁴ olor figure can be viewed at wileyonlinelibrary.com]

3.3 Preparation and characterization of INPs

The reaction between copperas solution and RPE synthesized produces a black solution as shown in Exhibit 4.

When RPE was reacted with a solution of copperas, the color of the solution changed rapidly to black from brown, and a precipitate formed as shown in Exhibit 4. The same event also occurred in the reaction of orange peel extract with $FeCl₃$ (Wei et al., [2016\)](#page-12-0), green tea extract, cloves, mint plants, pomegranate, and red wine with $FeCl₃$ and neem leaves with $FeSO₄$.7H₂O (Ravikumar et al., [2016\)](#page-11-0). This shows that there has been a reaction between Fe^{2+} ions from copperas with polyphenols from RPE to form INPs.

The formation of INPs from RPE can also be proven by observing the formation of SPI¹⁰ arface Plasmon Resonance). SPR is a resonance phenomenon between visible light waves and electrons on a metal surface that produces oscillations of electrons on the quantized metal surface. Exhibit 5 shows the absorption analysis spectra of the SPR peak at 214 nm absorption.

These results are similar to published studies at 214 nm (Harshiny et al., [2015\)](#page-10-0), 216 nm absorption, and 211 nm (Ravikumar et al., [2016\)](#page-11-0).

Polyphenols form INPs with the following steps: (a) formation of Fe complex compounds, (b) simultaneous Fe^{2+} ion reduction, and (c) oxidized capping. Thus, the mechanism of iron nanoparticle formation from rambutan peel extract is as follows:

(1) Formation of complex compounds

$$
nFe^{2+} + 2Ar - (OH) n \rightarrow n[Fe^{2+} - 2Ar = O] + 2nH^{+}
$$

(2) Simultaneous reduction of $Fe²⁺$

$$
n[Fe^{2+} - 2Ar = O]nFe^0 + 2nAr = O
$$

(3) Prevention of oxidation

EXHIBIT 5 UV-Vis SPR spectra of INPs resulting from the reaction of copperas with RPE. [Color figure can be viewed at wileyonlinelibrary.com]

$$
\frac{2}{2}(x+1)Fe^{0} + 2Ar = O + yO_{2} \rightarrow 2(Fe^{0} - FexOy - Ar = O)
$$
 (Wang et al., 2014).

In stages (1) and (2) of INP formation, polyphenols form complexes directly and then reduce Fe^{2+} ions (valence 2) to Fe^{0} (valence 0). The process of reduction and oxidation reactions depends on the reduction potential (E^0) of each reagent. According to Wang et al. [\(2017\)](#page-12-0), polyphenols have a reduction potential of 0.57 V sufficient to reduce Fe²⁺ to Fe⁰ which has a reduction potential of -0.44 V.

$$
Fe^{2+} + 2e \rightarrow Fe^{0} \tE^{0} = \frac{2}{2}44 \text{ V}
$$

$$
2Ar - (OH) n \rightarrow 2nAr = O + 2nH^{+} + 2e^{-C} = 0.57V
$$

+

$$
Fe^{2+} + 2Ar - (OH) n \rightarrow Fe^{0} + 2nAr = O + 2nH^{+} E_{Cell}^{0} = 0.13V
$$

A reaction can be said to be spontaneous if it meets the thermodynamic requirements, namely, its Gibbs free energy (ΔG°) < 0. The value of ΔG◦ can be determined from the standard cell potential by the

EXHIBIT 6 Elemental content test results of INPs.

formula:

 ΔG^0 = $-nFE^0_{Cell}$

Description: ∆G¹⁴, bbs free energy n, number of electrons involved,

F, Faraday's constant, 96500 C.mol−¹

If a cell has a positive E_{Cell}^0 price, then the ΔG° price will be negative and the reaction will be spontaneous. Therefore, the reaction between RPE and $Fe²⁺$ ions is spontaneous. This can be seen when the copperas and RPE solutions are mixed, there is an immediate reaction to form INPs (black solution) as shown in Exhibit [4.](#page-5-0) In general, the reaction mechanism for the synthesis of INPs wi[th po](#page-13-0)lyphenols is:

$nFe^{2+} + 2Ar - (OH) n \rightarrow nFe^{0} + 2nAr = O + 2nH^{+}$

Ar is a group of phenyl and n is the number of OH groups that reduce Fe2⁺ (Mystrioti et al., [2016;](#page-11-0) Wang et al., [2017\)](#page-12-0).

At stage (3), in the form of an RPE solution, capping and stabilization by polyphenols occur so that $Fe⁰$ is not oxidized. The presence of capping containing Fe, C, and O elements is also shown by SEM-EDX analysis results (Exhibits 6 and 7).

Exhibit [6](#page-7-0) shows intense peaks indicatin 2^8 e presence of Fe, C, and O. The elements involved in the formation of INPs are shown in Exhibit 7, namely, O (45.36%), C (25.32%), Fe (19.64%), N (7.68%), S (1.20%), P (0.63%), and K (0.17%). The presence of Fe, C, and O is a sign that INPs have been successfully synthesized from RPE. This result is similar to the published research b \int^2 ang et al. [\(2017\)](#page-12-0), Noruzi and Mousivand [\(2015\)](#page-11-0), and Ravikumar et al. [\(2016\)](#page-11-0).

SEM analysis results in the form of INPs as shown in Exhibit [8.](#page-7-0)

In Exhibit [8,](#page-7-0) it can be seen that most of the iron nanoparticles are round with a size of 10–70 nm. These results are similar to those of achado et al. [\(2013\)](#page-11-0), Poguberović et al. [\(2016\)](#page-11-0), and Wei et al. [\(2016\)](#page-12-0). Smaller particle size has a wider surface, are more reactive, and more higher reactivity. 2

The results of the XRD analysis of INPs are shown in Exhibit [9.](#page-7-0)

Exhibit [9](#page-7-0) shows that the solid nature of the synthesized INPs is amorphous. This is shown by the results of XRD analysis of INPs which did not show any diffraction peaks. A similar form was also reported

by Noruzi and Mousivand [\(2015\)](#page-11-0) who used fan fir extract and Shahwan et al. [\(2011\)](#page-11-0) who used green tea extract and rice bran extract. A similar form was also reported by Devatha et al. [\(2016\)](#page-10-0) who used leaf extract and Fazlzadeh et al. [\(2017\)](#page-10-0) who used plant extract.

The reaction between Fe^{2+} and polyphenols is shown in Exhibit [10.](#page-7-0) Exhibit [10](#page-7-0) shows the reaction between Fe^{2+} and polyphenols characterized by the presence of IR absorption by the OH group (phenol) in the 3471 cm⁻¹ region. In this area, there is a shift of peaks from 3471 cm−¹ to 3388 cm[−]1. The shift from 3471 to 3388 cm[−]¹ indicates the involvement of O-H functional groups. The peak shift from 1638 to 1628 cm⁻¹ indicates that C = C aromatic compounds are involved in the nanoparticle synthesis process. The peak shift from 1466 to 139⁰⁶ m⁻¹ indicates the involvement of the carboxyl group of RPE in the synthesis of INPs. The peak at 697 cm^{-1} corresponds to the C-H stretching of aromatic compounds.

Rambutan peel mainly contains pectin, cellulose, and hemicellulose as well as polyphenols . These polymers, especially polyphenols reduce Fe²⁺ to Fe⁰ (Kokila et al., [2015\)](#page-11-0). The reaction between RPE and copperas solution proceeds spontaneously to produce a black solid. RPE can reduce Fe^{2+} to Fe⁰ as seen from the brown solution turning black. The results of the characterization of INPs show that the compound contains $Fe⁰$ elements which are mostly spherical with a particle size of 10–70 nm. From these results, it can be concluded that the reaction between RPE and copperas solution produces INPs. The weight percentage of Fe synthesized by RPE is only 19.64%, but the use of rambutan peels can add value to unused waste that can cause environmental pollution.

3.4 Determination of efficiency and adsorption type of hexavalent chromium removal by INPs

a. The effect of pH

The effect of pH on hexavalent chromium removal was investigated from changes in pH of the solution of 3; 7 and 11 with different time intervals. The relationship between the initial pH of the solution and th ²² ercentage removal of hexavalent chromium is shown in Exhibit [11.](#page-8-0)

The percentage removal of hexavalent chromium can be seen in Exhibit [11,](#page-8-0) which increases as the pH decreases. The percentage removal of hexavalent chromium ions reached a maximum and remained relatively constant at 5 min at pH 3 and 20 min at pH 7 and 11. Among the various pH conditions, the maximum pH of removal was 3. At low pH (1-6), the dominant hexavalent chromium species is CrO_4^- and the surface of iron nanoparticles is positively charged . With increasing pH, there is a shift of $HCrO_4^-$ species to CrO_4^{2-} species which are stable at $pH > 6$.

Hexavalent chromium removal by INPs involve \mathcal{V} e reduction of hexavalent chromium to trivalent chromium and the oxidation of F^0 to Fe^{2+}/Fe^{3+} . In this reaction, electron transfer from Cr^{6+} to Fe⁰ occurs.

$$
HCrO4- + 7H+ + Fe0 \rightarrow Fe3+ + Cr3+ + 4H2O
$$

EXHIBIT 7 EDX spectrum of INPs. [Color figure can be viewed at wileyonlinelibrary.com]

EXHIBIT 8 SEM analysis of INPs. [Color figure can be viewed at wileyonlinelibrary.com]

$$
(1-x) Fe3+ + (x) Cr3+ + 2H2O + 3e \rightarrow Fe(1-x)CrxOOH(s) + 3H+
$$

Furthermore, 13 (III) precipitates as $Cr_xFe_{1-x}(OH)_3$ or $Cr_xFe_{1-x}(OOH)$, $x < 1$ (Poguberović et al., [2016\)](#page-11-0). So the lower the pH the greater the adsorption of INPs on hexavalent chromium.

b. Effect of hexavalent chromium concentration

The relationship between the initial concentration of hexavalent chromium and hexavalent chromium removal efficiency is shown in Exhibit [12.](#page-8-0)

Exhibit [12](#page-8-0) shows that 27 e removal efficiency of hexavalent chromium decreased as the initial concentration of hexavalent chromium increased from 5 to 15 ppm. The decrease in hexavalent chromium removal percentage is due to INPs having limited activity, which will become saturated at a certain concentration (Wang et al., [2014\)](#page-12-0). This result is similar to the results reported by Fazlzadeh et al.

EXHIBIT 9 Results of XRD analysis of INPs.

EXHIBIT 10 FTIR spectrum for INPs and RPE. [Color figure can be viewed at wileyonlinelibrary.com]

EXHIBIT 11 Effect of pH on hexavalent chromium removal with INPs. [Color figure can be viewed at wileyonlinelibrary.com]

EXHIBIT 12 Effect of hexavalent chromium concentration on hexavalent chromium removal. [Color figure can be viewed at wileyonlinelibrary.com]

[\(2017\)](#page-10-0) who used three plant extracts for hexavalent chromium removal and who used fertilizer production waste for hexavalent chromium removal. The percentage of hexavalent chromium removal decreased as the concentration of hexavalent chromium contaminant increased and showed high adsorption capacity of the synthesized INPs. The higher the hexavalent chromium concentration, the lower the percentage removal because INPs have a maximum limit in removing hexavalent chromium.

c. Effect of INP concentration

The results of the hexavalent chromium removal analysis with INPs varying the concentration of INPs are shown in Exhibit 13.

The hexavalent chromium removal percentage increased from 40.72% to 99.99% as the concentration of INPs increased from 0.01 to 0.15 g/100 mL. The hexavalent chromium removal percentage increases due to an increase in surface area location available for removal. Thus,²³ le percentage removal of hexavalent chromium is higher in proportion to the greater concentration of iron nanoparticles.

d. Effect of contact time

EXHIBIT 13 Effect of INPs concentration on hexavalent chromium removal with INPs. [Color figure can be viewed at wileyonlinelibrary.com]

EXHIBIT 14 Effect of contact time on hexavalent chromium removal with INPs. [Color figure can be viewed at wileyonlinelibrary.com]

The results of the analysis of hexavalent chromium removal with INP contact time variation are shown in Exhibit 14.

he percentage removal of hexavalent chromium increased from 39.53% to 41.31% as the contact time increased from 3 to 120 min for an initial hexavalent chromium concentration of 10 mg/L. The percentage removal was optimal at 10 min. The longer the contact time between INPs and hexavalent chromium, the more hexavalent chromium is adsorbed until a certain time limit. [12](#page-14-0)

Based on the results from points a, b, c, and d above, the removal of hexavalent chromium (Cr(VI)) using iron nanoparticles is an effective and promising method to treat contaminated water. It is commonly found in industrial wastewater, and its removal is crucial to ensure safe drinking water and protect the environment (Cheng et al., [2023;](#page-10-0) Prema et al., [2022\)](#page-11-0). INPs have gained attention as a potential remediation method due to their ability to reductor (VI) to less toxic and less mobile trivalent chromium (Cr(III)) through a process called chemical reduction (Yang et al., [2023\)](#page-12-0). Hexavalent chromium removal using INPs is generally effective, but it is necessary to consider certain factors, such as pH, temperature, and the presence of other contaminants, which can affect the efficiency of the process. Overal²⁵ e use of INPs for

EXHIBIT 15 Results of isotherm type determination of hexavalent chromium metal by INPs.

$C_{Cr(VI)$ Early (ppm)	C_e (ppm)	Qe (mg/g)	C_e /qe	Log qe	Log C _e
	0.15	4.85	0.031	0.686	-0.824
10	0.32	9.68	0.033	0.986	-0.495
15	0.52	14.48	0.036	1.161	-0.284
20	0.73	19.27	0.038	1.285	-0.137
25	0.95	24.05	1.381	1.381	-0.022

hexavalent chromium removal is a promising and effective technology (Garvasis et al., [2023;](#page-10-0) Rajapaksha et al., [2022\)](#page-11-0).

e. Determination of adsorption isotherm type

Determination of adsorption isotherm type was done by entering equilibrium data into Equation [\(4\)](#page-3-0) (Langmuir isotherm) and Equation (5) (Freundlich isotherm), the results of which are shown in Exhibit 15.

If a straight line is made using the Langmuir isotherm equation, the Ce/qe versus Ce curve, and the Freundlich isotherm equation, the log Ce versus log qe curve will be obtained as shown in Exhibits 16 and [17.](#page-10-0)

Exhibits 16 and [17](#page-10-0) show that the adsorption equation of hexavalent chromium ions by INPs fulfill.¹⁸ le Langmuir isotherm equation with $R^2 = 0.9833$ and the Freundlich isotherm equation with $R^2 = 0.9995$. This indicates that Langmuir and Freundlich's equations can b^{26} plied to the adsorption process of hexavalent chromium by INPs (Rahmani et al., 2011; Khatoon et al., 2013). The correlation coefficient (R2) of Freundlich isotherm (R² = 0.9995,¹⁶) more suitable than the Langmuir model ($R^2 = 0.9833$) for hexavalent chromium adsorption. This means that the Freundlich isotherm model is more suitable than the Langmuir isotherm model. This result is similar to the study δ^9 oguberović et al. [\(2016\)](#page-11-0) on the adsorption of As(III) and hexavalent chromium using INPs from oak, mulberry, and cherry leaf extracts which fit the Freundlich isotherm model. From the Langmuir isotherm straight line equation, $y = 0.0109 x + 0.0296$, the adsorption capacity (qmax) of

INPs on hexavalent chromium can be determined from the intercept price $1/q$ max = 0.0296, so that the adsorption capacity price, qmax is 91.74 mg/g hexavalent chromium. Based on the price of R^2 , the adsorption process between INPs and hexavalent chromium takes place in a multilayer manner.

4 CONCLUSION

The conclusions of this research are:

- 1. The level of copperas produced from iron scrap in the lathe workshop has a Fe content of 19.84 \pm 0.15%. These results meet SNI 06-4888-1998 standards regarding copperas quality requirements with a minimum Fe content of 19%.
- 2.4 ambutan peel extract contains phenol compounds with a total phenol content of 877.39 \pm 16.6 ppm/100 g mg/100 g rambutan peel. This result is equivalent to 441.42 mg GAE/100 g rambutan peel.
- 3. The characteristics of iron nanoparticles produced from the reaction of rambutan peel extract with copperas from iron scrap of the lathe workshop are on average spherical with a size of 10° am.
- 4. Hexavalent chromium removal using INPs was maximum at pH 3.1, hexavalent chromium concentration of 5 ppm, INPs concentration of 0.10 g/100 mL, and contact time of 10 min. The adsorption

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EXHIBIT 17 Freundlich isotherm curve of hexavalent chromium adsorption by INPs. [Color figure can be viewed at wileyonlinelibrary.com]

 1 otherm type of hexavalent chromium using INPs can take place chemically or physically. [The](#page-13-0) correlation coefficient showed that the Freundlich model (R² \sim 99) was more suitable than the Langmuir model ($R^2 = 0.98$) with an adsorption capacity of 91.74 mg INPs/g hexavalent chromium. Overall, the use of INPs for hexavalent chromium removal is a promising and effective technology.

CONFLICT OF INTEREST STATEMENT

The authors declare that they have no competing interests.

DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available in repository .setiabudi.ac.id at [http://repository.setiabudi.ac.id/cgi/](http://repository.setiabudi.ac.id/cgi/users/home) [users/home.](http://repository.setiabudi.ac.id/cgi/users/home) These data were derived from the following resources available in the public domain: [list resources and URLs].

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competing interests.DATA AVAILABILITY STATEMENTThe data that support the fi...

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