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ORIGINAL RESEARCH PAPER

Machine learning using random forest to model heavy metals removal efficiency using a zeolite-embedded sheet in water

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ABSTRACT

BACKGROUND AND OBJECTIVES: Zeolite has been recognized as a potential adsorbent for heavy metals in water. The form of zeolite that is generally available in powder has challenged the use of zeolite in the environment. Embedding powder zeolite in a nonwoven sheet, known as a zeolite-embedded sheet can be an alternative to solve that. Another challenge is that information and models of zeolite-embedded sheet removal efficiency are still limited. The novelty of this study is, first, the development of a zeolite-embedded sheet to remove heavy metals from water, and second, the use of the random forest method to model the heavy metal removal efficiency of a zeolite-embedded sheet in water.

METHODS: The heavy metals studied were copper, lead and zinc, considering that those are common heavy metals found in water. For developing the zeolite-embedded sheet, the methods include fabrication of the zeolite-embedded sheet using a heating procedure and heavy metals adsorption treatment using the zeolite-embedded sheet. The machine learning analysis to model the heavy metal removal efficiency using zeolite-embedded sheet was performed using the random forest method. The random forest models were then validated using the root mean square error, mean square of residuals, percentage variable explained and graphs depicting out-of-bag error of a random forest.

FINDINGS: The results show the heavy metal removal efficiency was 5.51-95.6 percent, 42.71-98.92 percent and 13.39-95.97 percent for copper, lead and zinc, respectively. Heavy metals were reduced to 50 percent at metal concentrations of 10.355 milligram per liter for copper, 171.615 milligram per liter for lead and 4.755 milligram per liter for zinc. Based on the random forest models, the important variables affecting copper removal efficiency using zeolite-embedded sheet were its contents in water, followed by water temperature and potential of hydrogen. Conversely, lead and zinc removal efficiency was influenced mostly by potential of hydrogen. The random forest model also confirms that the high efficiency of heavy metals removal (>60 percent) will be achieved at water potential of hydrogen ranges of 4.94–5.61 and temperatures equal to 29.1 degrees Celsius.

CONCLUSION: In general, a zeolite-embedded sheet can adsorb diluted heavy metals from water because there are percentages of adsorbed heavy metals. The random forest model is very useful to provide information and determine the threshold of heavy metal contents, water potential of hydrogen and temperature to optimize the heavy metal removal efficiency using a zeolite-embedded sheet and reducing pollutants in the environment.

NUMBER OF REFERENCES	NUMBER OF FIGURES	NUMBER OF TABLES
62	10	5

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INTRODUCTION

Water is a basic necessity for humans (Corbella, 2010; Daneshvar Rad *et al.,* 2023; Sarmurzina *et al.,* 2023). Water sources for human consumption include well (Schaider *et al.,* 2016), rainwater collection (Evantri *et al.,* 2021), and surface water that service providers treat (Dinh et al., 2020). Water that meets health requirements must be safe in terms of physical, chemical, microbiological, and radioactive aspects (WHO, 2022). Heavy metals are naturally occurring environmental components and are considered pollutants in high concentrations (Sabiha-Javied *et al.,* 2009; Astuti *et al.,* 2021; Ehzari *et al.,* 2022; Sulistyowati *et al.,* 2023a). The higher the pollution of heavy metals in a body of water, the higher the bioaccumulation of heavy metals in the tissues of aquatic organisms (Junianto *et al.,* 2017; Safari *et al.,* 2019; Nurhasanah *et al.,* 2023; Sulistyowati *et al.,* 2023b). The increasing human population and its anthropogenic activities affect the supply of groundwater and surface water contaminated with heavy metals, causing disruptions to the balance of organisms and aquatic biota (Waqar *et al.,* 2013; Fahimah *et al.,* 2023; Sabilillah *et al.,* 2023). These may disturb the ecological balance of the environment and the diversity of aquatic organisms (Budijono and Hasbi, 2021). Several methods for removing heavy metals from water have been developed, including chemical precipitation (Djedidi *et al.,* 2009), ion exchange (Al-Othman *et al.,* 2011), reverse osmosis (Aljendeel, 2011) and membrane separation (O'Connel *et al.,* 2008). These methods produced encouraging results. The metal removal efficiency and dewatering ability of the resulting sludge were assessed using studies of simple and combined precipitation treatment modes (Djedidi *et al.,* 2009), physiochemical properties and higher ion exchange capacity of composite cation exchangers, which improves the efficiency of toxic metal separation, adsorption and removal (Al-Othman *et al.,* 2011). Metals are efficiently separated by reverse osmosis (Aljendeel, 2011) and the membrane separation approach uses cellulose as the foundation for building new adsorbents that are reasonably straightforward to chemically change (O'Connel *et al.,* 2008). However, these technologies are too expensive for treating and disposing secondary toxic metal sludge, take too long to treat, consume too much energy or are ineffective when heavy metals are present in the wastewater at low concentrations (Kabwadza-Corner *et al.,* 2015). Researchers are investigating low-cost adsorbents for heavy metal removal, such as chitosan beads (Ngah *et al.,* 2006), alginate beads (Samimi and Moeini, 2020), sago waste (Quek *et al.,* 1998), rice husks (Wong *et al.,* 2003), bacterial biomass (Samimi and Shahriari Moghadam, 2021), sawdust (Larous *et al.,* 2005) and zeolites (Peric , 2004). One of the potential adsorbents is zeolite, which contains silicon (Si), aluminum (Al) and oxygen (O) atoms. The primary roles of zeolite are ion exchange or as an adsorbent, molecular sieve, catalyst and soil amendment. Because zeolite and clays contain negative charges, zeolite can absorb heavy metals or positively charge dyes, including textile waste. Zeolites are efficient adsorbent materials with numerous uses in the removal of heavy metals from wastewater. Zeolite was synthesized from by-products such as fly ash and rice husk. The sorption process of zeolites was generally spontaneous and endothermic, and heavy metal ions were removed by adsorption and ion exchange processes. Sadia *et al.* (2021) found that zeolites have good cation exchange capabilities and sorption performance. One difficulty in using zeolites for metal removal is that, despite their strong adsorptive capacity for heavy metals, recovering zeolites after adsorption is challenging because zeolites are in powder form. Then, a solution is required to use other zeolite form in water, one of them is creating a zeolite-embedded sheet (ZES). This method is proposed as an effective adsorbent for heavy metals, primarily in water. The advantage of embedding powder zeolite in a nonwoven sheet is the convenience with which an adsorbent can be collected after adsorption, and they require instruments to separate the adsorbent and adsorbate. Botoman *et al.* (2018) demonstrated the efficacy of a Linde type A (LTA)-embedded sheet in eliminating lead (Pb) from water. Random forest (RF) methods have been widely used, including in heavy metal studies. Tan *et al.* (2018) have explored heavy metal estimation of soil using RF due to its built-in modeling expressiveness and feature selection ability. RF has also been used to estimate, validate and model the heavy metal removal efficiency from environments. Currently, advanced data analysis approaches, including machine learning (ML), have become versatile tools for modeling and developing estimation and prediction of the future of unknown data based on hidden information related to massive input data. In heavy metal removal studies, RF has been used to accurately predict heavy metal adsorption efficiency using biochar (Zhu *et al.*, 2019). According to Chun *et al.* (2022), the heavy metal removal efficiency based on flocculant properties, flocculation conditions and heavy metal properties can be predicted using RF with high accuracy with a coefficient of determinant (R^2) equal to 0.9354. In Indonesia, estimating the efficiency of heavy metal removal from water using both ZES and RF analyses is still lacking and accurate information on the heavy metal removal efficiency of ZES is needed because of the rising heavy metal contents in water. Considering this situation, this study attempts to address the following questions: Can ZES remove heavy metals like copper (Cu), lead (Pb) and zinc (Zn) from water? And if ZES can remove heavy metals, can RF model the removal efficiency of ZES? The objectives of this study are to assess the Cu, Pb and Zn removal efficiency of ZES and model the removal efficiency using RF. The result of this study can contribute to heavy metal removal practices, in particular by providing information on the heavy metal concentration at which ZES can be used and providing the optimum removals. This study has been conducted at the Department of Biology and Chemistry, Faculty of Mathematics and Natural Sciences, Universitas Indonesia in 2023.

MATERIALS AND METHODS

Materials

Chemical reagents for adsorption testing, namely copper (II) sulfate pentahydrate ($Cu(SO_4)_2.5H_2O$), zinc sulfate heptahydrate ($ZnSO_4.7H_2O$), lead (II) nitrate ($Pb(NO_3)_2$), sodium nitrate ($NaNO_3$) and sodium sulfate (Na_2SO_4), were ordered from MERCK. Aquades was bought directly from Brataco, limited (Ltd), Indonesia. Raw materials were purchased from different marketplace. Nonwoven sheets made of polypropylene (PP) and polyethylene (PE) were bought from Platec, Ltd, Japan. The zeolite A-4 powder was acquired from Wako Pure Chemical Industries, Ltd, Japan.

Fabrication of ZES

In this study, ZES was fabricated, referring to Sadia *et al.* (2021). The nonwoven sheets were cut into 81 centimeter squared (cm^2) and divided into nine pieces. Each of them was then numbered and weighed. The amount of zeolite powder was poured into a stainless tray. The sheets were placed into the tray, and then pressed slowly and inverted by hand. The sheets were then heated at 160 °C for 8 minutes. After chilling, ten embedded sheets were put into a 1 liter (L) Duran bottle, added with 500 milliliter (mL) of water and shaken by hand for 2 minutes. After the water is removed, this shuffling is repeated one more time. Then, the sheet in the bottle is shaken with 500 mL water for 1 hour to remove zeolite powder that does not attach to the filter. Shaking was repeated once, and the filter was dried at room temperature (25 °C). The dry weight of the sheet was weighed to determine the mass of zeolite contained in each embedded sheet.

Characterization of ZES

ZES was recorded for their structure, chemical composition, and vibrational properties. A scanning electron microscope–energy-dispersive X-ray (SEM-EDX) (Jeol JSM-IT200) was used to observe the surface morphology and chemical properties of ZES. Fourier transform infrared (FTIR) (Thermo Scientific Nicolet iS50 FTIR) was applied for vibrational properties. SEM-EDX analysis was conducted in the National Research and Innovation Agency (BRIN) and the FTIR was conducted in the Integrated Laboratory and Research Center (ILRC), University of Indonesia.

Cu, Pb, and Zn adsorption using ZES

Adsorption solutions were prepared with different concentrations. First, 1 molar (M) $NaNO_3$ and Na_2SO_4 solution as a background solution were composed. Second, stock solutions of 0.1 M $Cu(SO_4)_2.5H_2O$ (1000 mL), 0.1 M $Pb(NO_3)_2$ (1000 mL) and 0.1 M $ZnSO_4.7H_2O$ (1000 mL) were prepared. Third, stock solutions of 5 millimolar (mM) Cu, 5 mM Pb and 5 mM Zn were then prepared. The Cu, Pb and Zn solutions were diluted from various concentrations to experiment with the adsorption of Cu, Pb and Zn solutions with ZES. For $Cu(SO_4)_2.5H_2O$, $Pb(NO3)_2$ and $ZnSO_4.7H_2O$ 50 mL solution at each concentration: 0.025, 0.05, 0.10, 0.20, 0.,40 and 0.60 mM were taken for initial concentration analysis using the Inductively Coupled Plasma (ICP) method and the potential of hydrogen (pH) was measured. In adsorption experiments, $Cu(SO_4)_2.5H_2O$, $Pb(NO3)_2$ and $ZnSO_4.7H_2O$ solutions were put into a PP bottle according to the weight of ZES, with ZES-solution ratio of 0.1 gram (g): 500 g.

Table 1: Summary of initial heavy metal contents in water

Prepared solution (mM)	Contents in water (mg/L)		
	Cu	Pb	Zn
0.025	1.365	6.765	1.575
0.05	2.7	9.38	3.31
0.1	5.685	28.035	6.44
0.15	8.3	30.92	4.755
0.2	10.355	38.895	6.11
0.3	14.84	74.62	4.67
0.4	19.395	100.515	26.95
0.5	21.015	139.01	34.24
0.6	28.26	171.615	41.125

One sheet of ZES was put in each bottle and shaken vertically 10 times. The bottle was chilled for 3 hours and shaken every 1 hour. Then, the ZES in the bottle is taken with tweezers. Finally, the pH of the solution was measured again and the final concentration of Cu in the solution was measured using ICP. The same treatment was also conducted on Zn and Pb adsorption. The heavy metal content analyses were performed at Saraswanti Indo Genetech Ltd, Indonesia. A summary initial heavy metal contents in water is shown in Table 1.

Environmental variables

In this experiment, two water environmental variables were measured. Those variables were water pH and temperature. The equipment used to measure the these variables was a Lutron pH meter 5510.

Cu, Pb, and Zn adsorption experiment

Heavy metal removal of ZES was measured on the basis of the remaining heavy metal contents after treatment with ZES. The percent removal of heavy metals was calculated using Eq. 1 (Chibuzo *et al.*, 2016; Azimi *et al*., 2019).

$$\%\text{adsorption} = \frac{C_i - C_e}{C_i} \text{ x } 100\ (\%) \qquad (1)$$

Where, C_i and C_e are the initial and final concentration of heavy metals, respectively.

RF model statistical analysis

The statistical analysis used in this study is RF. The RF model was developed following Fathi *et al.* (2014). An RF is a collection of hundreds of decision trees with identical distribution. Classification algorithms, such as classification and regression trees, are used to create these trees. RF, proposed by Leo Breiman, constructs many decision trees and blends them to obtain a more accurate and consistent prediction. This model, in terms of the strength of the individual predictors and their relationships, provides insight into the RF's capacity to forecast. The predictors in this study were water pH, temperature and heavy metal content in mM and milligram per liter (mg/L). Within RF, a classification tree is iteratively defined by a division criterion (node) obtained from one of the variables, x_j, which results in the construction of two subsets in the training sample consisting of a subset that contains the observations (i) that satisfy the condition x^i_j < a real number, which is defined by the algorithm (T), whereas the other subset contains the observations i that satisfy the condition $x^i_j > T$ (Ruiz-Gazen and Villa, 2008). For both classification and regression models, we utilized the RF package in R platform version 3.6.3 for statistical computing and making visuals. The reference contains a full description of the RF method.

Model validation for error analysis

The removal efficiency model developed using RF was validated and measured for its error. The validation was based on several statistical tests. Those tests include the root mean square error (RMSE), mean square of residuals (MSR), percentage variable explained and graphs depicting the out-of-bag (OOB) error of an RF model for Cu, Pb, and Zn. The lowest RMSE closed to zero (Sang *et al.*, 2022) and the more trees within the OOB mean the model is providing the best accuracy.

RESULTS AND DISCUSSION

Characterization of ZES

Fig. 1 shows the appearance of ZES morphology using SEM and the presence of zeolite - a cubical structure attached to sheet fibers. The sticking of zeolite occurs because of heating. The chemical composition of ZES analyzed using EDX is shown in Table 2. ZES in the study contained elements carbon (C) (66.86%), O (26.87%), natrium (Na) (2.56%), Al (1.75%), Si (1.96%) and chloride (Cl) (0.28%). The presence of O, Na, Al and Si indicated that zeolite is successfully attached to the sheet. Related to adsorption ability, silicon per aluminum (Si/Al) ratio is considered important. In this study, the Si/Al ratio present in the ZES is 0.96. According to Hudcová *et al*. (2021), the smaller the value of the ratio, the better the adsorption ability of a material. Because the lower in Si/Al ratio of the zeolite indicates the higher in its CEC (cation exchange capacity).

The vibrational properties of ZES using FTIR are shown in Fig. 2. The peak band at 3381.33 reciprocal centimeter (/cm) and 3643.07/cm indicate oxygen hydrogen (OH) stretch as reported by Corona *et al.* (2009) and Jacas-Rodríguez *et al.* (2020) stated that OH stretch usually detected in the range 3400–3700/cm that has interaction between the water hydroxyl and cations. Spectral regions between 3000/cm and 2800/cm enable an analysis of the peak on 2848.33/cm and 2915.36/cm related to the asymmetric and symmetric stretching vibrations of carbon hydrogen (C–H) bonds in the methoxy ($O–CH_3$) group (Portaccio *et al.,* 2011). C–H bending was observed at the peak of 1479.63/cm (Merck, 2023). The vibration at 969.69/cm is attributed to the asymmetric stretching vibrations characteristic of asymmetrical stretching vibrations (V_{as}) siloxane (Si–O–Si) and V_{as} silicon–

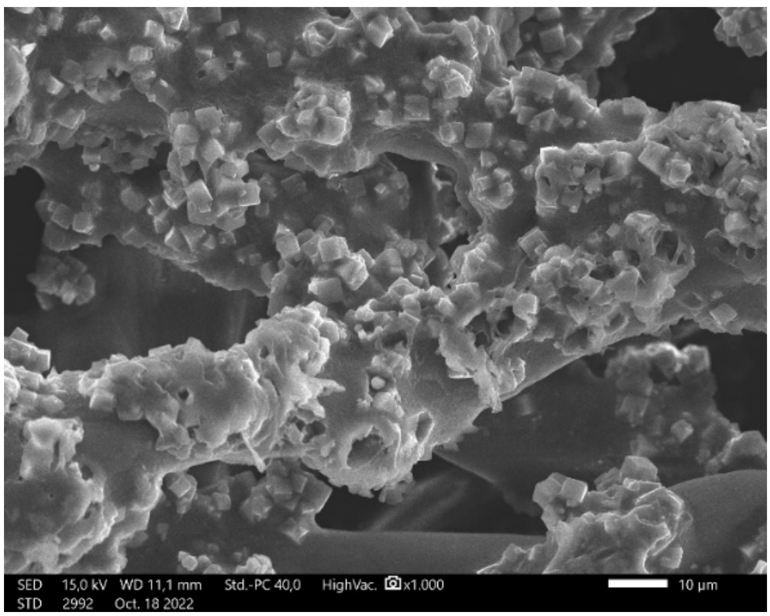

Fig. 1: Morphological structure of ZES

Table 2: Chemical composition of ZES

Element	Atomic concentration (%)
C	66.86
O	26.87
Na	2.56
Al	1.75
Si	1.69
Cl	0.28

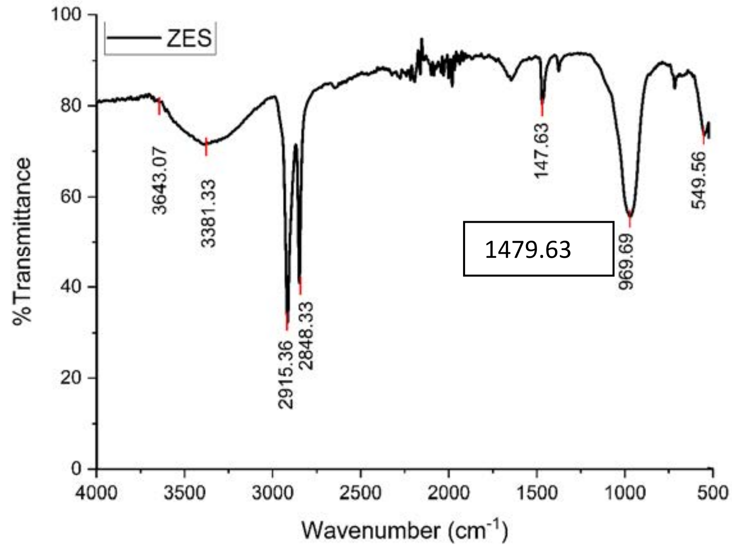

Fig. 2: FTIR spectrum of ZES

Table 3: Summary of model validation for Cu, Pb and Zn

Variables	Cu	Pb	Zn
RMSE	1.805	1.080	2.229
MSR	0.383	2.202	0.674
% var explained	99.09	99.33	99.92

oxygen–aluminum (Si–O–Al) (Mozgawa *et al*., 2011). The peak of 549.56/cm has a complex band as the superposition of different bands, composed of the symmetrical stretching vibrations (V_s) of Si–O–Si and the bending vibrations (δ) corresponding to siloxane (O–Si–O) (Jacas-Rodríguez *et al.,* 2020). The peaks related to the carbon hydrogen (C–H) bonds (2848.33 and 2915.36/cm) and the C–H bending (1479.63/cm) were due to PE (Madhu *et al.*, 2014) and PP (Ummartyotin and Pechyen, 2016) respectively, derived from the sheet. The other peaks were belong to zeolite.

Adsorption trends of Cu, Pb, and Zn

The adsorption trends and removal efficiency of Cu, Pb and Zn are shown in Figs. 3–5 after 60 minute contact times. In general, ZES can adsorb diluted heavy metals from water because percentages of adsorbed heavy metals are present. The adsorptive effects of this experiment were influenced by the initial heavy metal contents in water. It is clear that an increase in heavy metal contents in water will reduce heavy metal adsorption of ZES. The heavy metal removal efficiency of ZES was reduced to 50 percent (%) at metal concentrations of 10.355 mg/L for Cu, 171.615 mg/L for Pb and 4.755 mg/L for Zn. It can be concluded that ZES was more efficient at adsorbing Pb because the Pb removal efficiency has a threshold as high as 171.615 mg/L. The adsorption trends of Cu, Pb, and Zn were related to cation exchange capacity and porous sturcture. Solid materials with a porous structure can be used as adsorbents (Irani *et al.*, 2011). Zeolite has high adsorption efficiency due to its low cation exchange capacity in the form of high Si/Al ratios. These properties explain the adsorption result of zeolite and its potency as an adsorbent for those heavy metals in water. On the basis of Pratama *et al.* (2021), zeolite particles contributed Cu and Zn adsorption sites. Copper ion (Cu^{2+}) form complex with silicon–oxygen bonds (Si–O) and aluminum–oxygen bonds (Al–O). According to Chang and Shih (2000), zeolite's features and the characteristics of those metal ions impact the variation in adsorption capacity. Metal ions may pass through pores that have a particular size. The adsorption capacity would be reduced if a metal ion was larger than the pore size. Compared with Cu, Zn has a larger atomic diameter (Barak and Helmke, 1993). These characteristics explain the higher levels of Cu adsorption in the zeolite than Zn adsorption. More central metal ions have low capacities for solidity and electrostatic adsorptive power, these will restrict the ability of metals of particular dimensions to interact with one another (Minceva *et al.*, 2007). On the other hand, ions having a greater ion valence and a small ion radius will be firmly and densely adsorbed. According to Minceva *et al.* (2007), metals with higher electronegativity values will be simpler

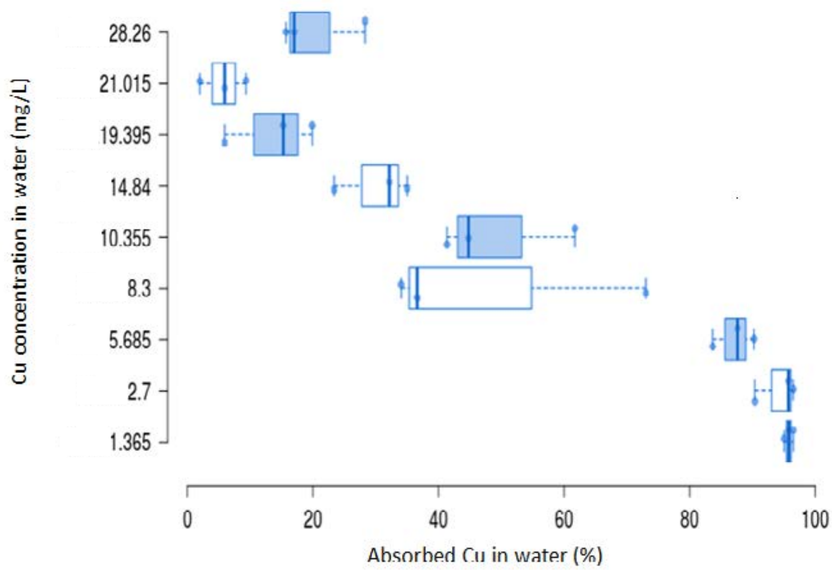

Fig. 3: Percentages of adsorbed Cu (x-axis) in water related to Cu concentration (y-axis; 1.365, 2.7, 5.685, 8.3, 10.355, 14.84, 19.395, 21.015, 28.26 mg/L) (means between population / distribution (F) = 87.999, probability (P) = 0.000)

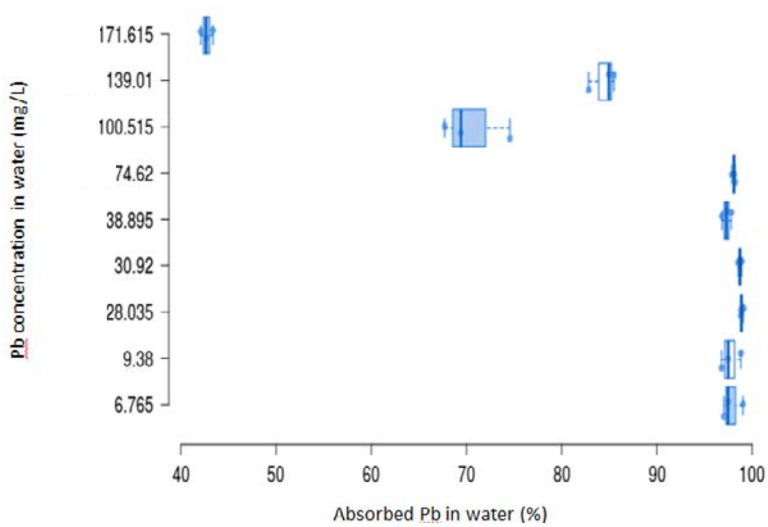

Fig. 4: Percentages of adsorbed Pb (x-axis) in water related to Pb concentrations (y-axis; 6.765, 9.38, 28.035, 30.92, 38.895, 74.62, 100.515, 139.01, 171.615 mg/L) (F = 570.797, P = 0.000)

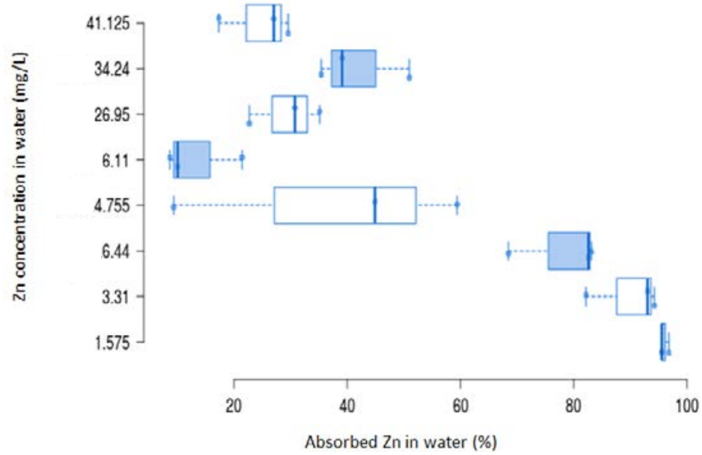

Fig. 5: Percentages of adsorbed Zn (x-axis) in water related to Cu concentrations (y-axis; 1.575, 3.31, 6.44, 4.755, 6.11, 26.95, 34.24, 41.125 mg/L) (F = 24.954, P = 0.000)

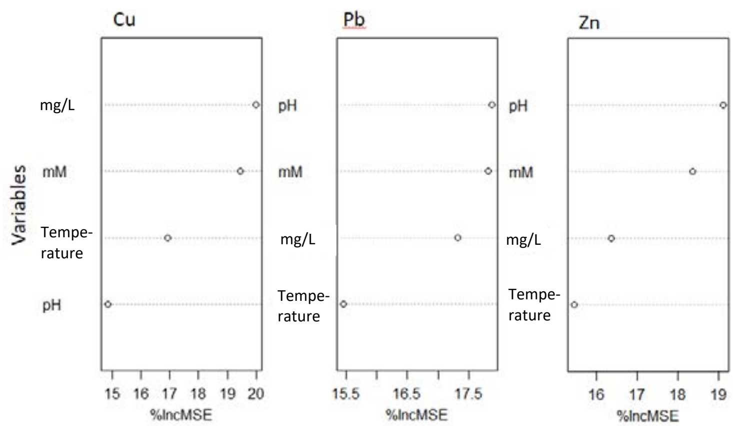

Fig. 6: Important variables for Cu, Pb and Zn adsorption by ZES

to absorb than those with lower electronegativity values. Additionally, zeolite displayed a typical silicon dioxide (SiO_2) and aluminum oxide (AlO_3) bond structure. Zeolite contains an extra negative ion capacity due to this connection, which is exploited in a cation exchange process to bind the metal ions. Our findings show that at metal concentrations such as 10.355 mg/L for Cu and 4.755 mg/L for Zn, heavy metals decreased to 50%. This makes ZES more efficient at adsorbing Cu ions because it can adsorb a higher concentration of 10.355 mg/L than Zn. If Zn ceoncentrations were close to 10.355 mg/L, the removal efficiency of ZES was only <50%. Cu is more highly adsorbed in zeolite than Zn. Cu can be highly adsorbed in zeolite because Cu has a smaller atomic size than Zn. Zn has a larger electron number than Cu, so the size of the Zn ion is larger than that of the Cu ion.

Important variables affecting the efficiency of removing Cu, Pb and Zn

Fig. 6 presents the important variables for Cu, Pb and Zn adsorption by ZES The important variables affecting Cu removal efficiency by ZES were its contents in water, followed by water temperature and pH. In the other hand, for Pb and Zn, removal efficiency was influenced mostly by pH. According to Kulkarni *et al*. (2013), pH has an influence on metal adsorption using zeolite because pH affects the H^+ between adsorbents and metals (Mubarak *et al*., 2022).

Besides pH, temperature also contributes to metal sorption by zeolite (Chibuzo *et al*., 2016).

Model validation for Cu, Pb, and Zn

Model validation for Cu, Pb, and Zn are shown in Table 3. Zn has the highest RMSE, whereas Pb has the lowest RMSE. Lower RMSE means better accuracy. Then the prediction accuracy of the model is good in this case of Pb and Zn.

OOB error

The OOB error for Cu, Pb, and Zn is shown in Fig. 7. All OOB shows a large number of trees when the OOB error is decreasing, which indicates the model has good accuracy. Between heavy metals, the best model was observed for Zn and Cu, followed by Pb. The number of trees in Zn and Cu had already stabilized at 300 and 400, respectively. Conversely the number of trees in Pb was still fluctuating after 400.

RF model

The RF model (Fig. 8) confirms that the high removal efficiency of Cu from water with a probability of 100% (>66%, average efficiency = 92.2%; confidence intervals (CI): 87%–97.4%) was significantly determined by the Cu(II) solution in water ($P < 0.001$), equal to ≤5.685 mg/L. While with a Cu(II) solution of >5.685 mg/L in water, ZES can only remove <66% (average efficiency = 25.5%; CI: 12.7%–38.3%) of Cu from water. On the basis of the RF model, Pb and Zn removal efficiency was affected by pH. A high removal efficiency of Pb from water with a probability of 100% (Fig. 9) was significantly determined by water pH equal to ≤4.94 and water temperature equal to ≤29.1 degrees Celsius (°C) ($P < 0.001$). When water pH equals to >4.94 and water temperature equals to >29.1 °C, ZES can only adsorb 30–60% of Pb from water. The Zn adsorption pattern was also similar to that of Pb adsorption. Based on the RF model for Zn (Fig. 10), the high removal efficiency of Zn from water with a probability of 100% was significantly determined by water pH equal to ≤5.61 ($P < 0.001$). With a high water pH equal to >5.61 and water temperature equal to >29.6 °C, ZES can only adsorb 30–60% of Zn from water. In this study, several heavy metal removal efficiency has been modeled using RF. According to Shi *et al.* (2022), ML methods have been used in heavy metal content assessment studies that include artificial neural networks (ANN) (Sakizadeh *et al.*, 2017), least absolute shrinkage and selection operators (LASSO), genetic algorithms (GA), and error back-propagation neural networks (BPNN), namely the LASSO-GA-BPNN model, support vector regression (SVR) (Huang *et al.*, 2021) and RF (Taghizadeh-Mehrjardi *et al.*, 2021). The RF model used in this study has obvious high accuracy and recognition capability for predicting the removal efficiency of heavy metals from water. The results also show that the RF model is effective at predicting adsorbed Cu, Pb and Zn, confirming the science and the advancement of the RF prediction model in heavy metal removal studies (Cao *et al.*, 2023). According to the model, removal efficiency was also affected by water pH and temperature, as can be seen for Pb and Zn. In this study, high removal efficiency was observed when the pH was <4.94 for Pb and 5.61 for Zn. This finding contradicts the general pattern. Shaker (2007) confirms that an increase in pH will increase adsorption

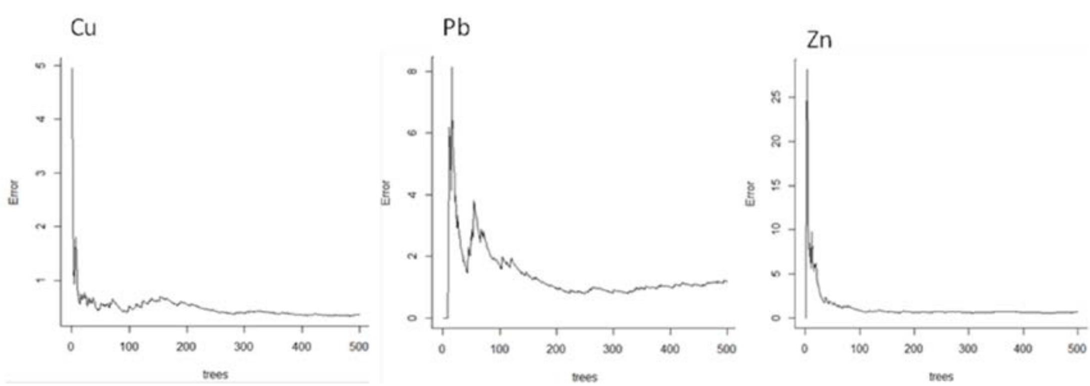

Fig. 7: Graphs depicting the OOB error of an RF model for Cu, Pb and Zn

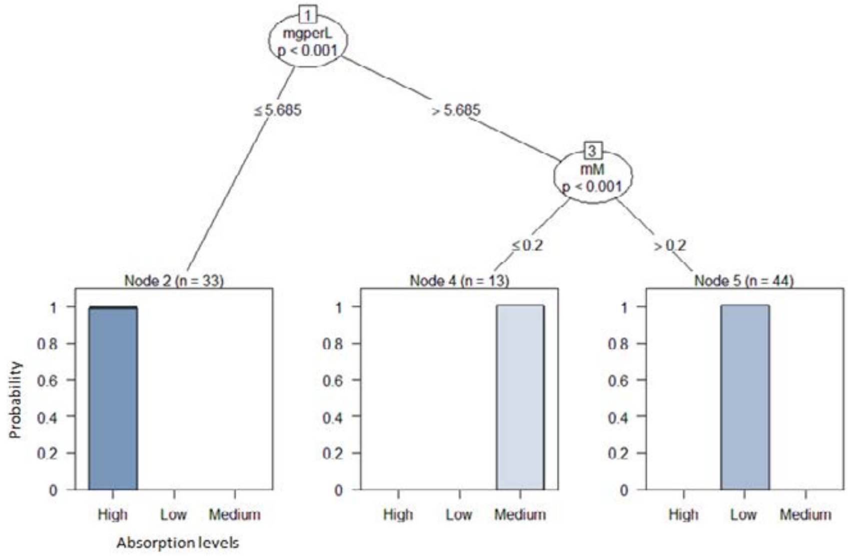

Fig. 8: RF model of adsorbed Cu (x-axis, abosption levels: low, 0–30%; medium, 31–60% and high, 61–100%) in water related to water pH, temperature and Cu concentration in mM and mg/L.

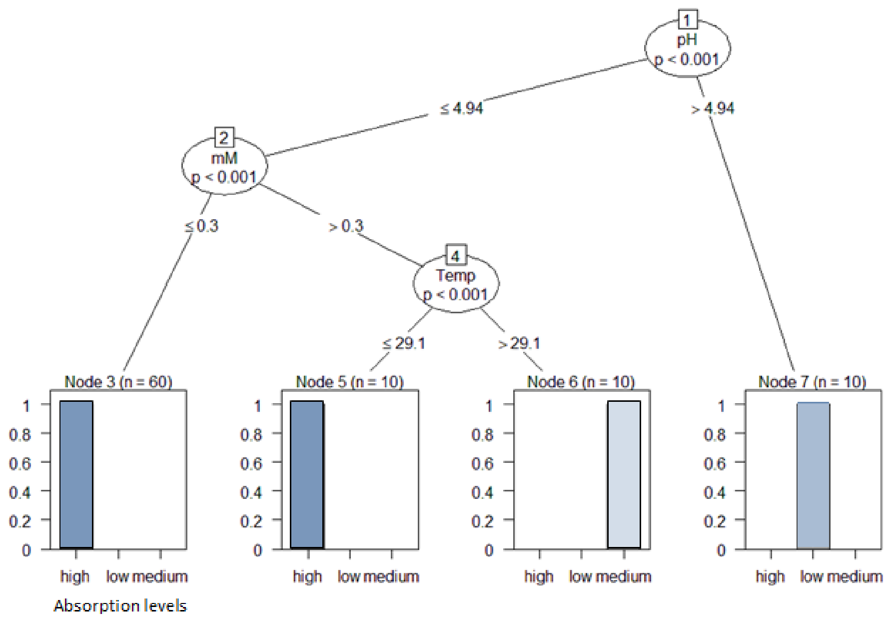

Fig. 9: RF model of adsorbed Pb (x-axis, absorption levels: low, 0–30%; medium, 31–60% and high, 61–100%) in water related to water pH, temperature and Pb concentration in mM and mg/L.

since, at low pH, there is a high concentration of H^+ with high mobility due to metal ions, and this increases the competition between H^+ and metal ions, causing reductions in its adsorption. The removal efficiency using ZES in this study was comparable to the previous study (Table 4). For zeolite adsorption, an increase in pH will reduce the adsorption. Kulkarni *et al*. (2013) observed that when pH increased from 6 to 8, the

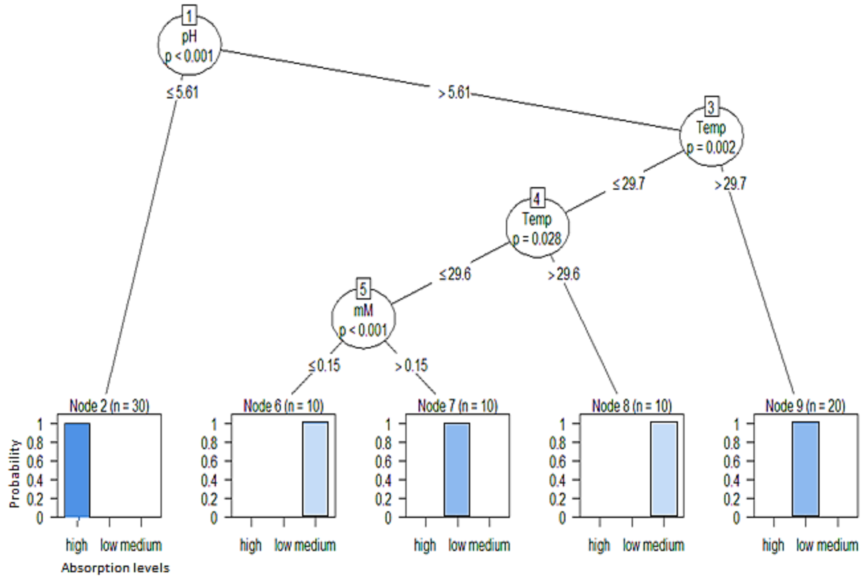

Fig. 10: RF model of adsorbed Zn (x axis, abosption levels: low, 0–30%; medium, 31–60% and high, 61–100%) in water related to water pH, temperature and Zn concentration in mM and mg/L.

Table 4: Comparisons with previous studies and other zeolite adsorbents

Heavy metals	Adsorbents	Removal efficiency (%)	pH ranges	Temperature (^{O}C)	Sources
Calcium (Ca)	Zeolite 4A	70–90	3–8	NA	Kulkarni *et al.* (2013)
iron (Fe) and manganese (Mn)	Titanium dioxide (TiO_2)@Zeolites-4A nanocomposite	70–100	2–9	NA	Mubarak *et al.* (2022)
Pb	Zeolite solution	80–100	2–10	29.8–39.8	Chibuzo *et al.* (2016)
Cu, Pb and Zn	ZES	5.51–98.92	4–7	29.1–29.9	This study

removal efficiency decreased from 90% to 70%. Even at a low pH of 2 to 3, the removal efficiency of zeolite can reach 70-80%. Temperature was also considered an important variable for removing heavy metals from water. In this study, adsorptions was observed at temperatures of 29.1–29.9 °C. This temperature range is in agreement with that in previous studies. Chibuzo *et al.* (2016) reported that 29 °C is the optimum temperature for zeolite. Instead, although this study was successful in predicting metal adsorption, the RF approach still has significant shortcomings that must be addressed (Fernández-Delgado *et al.*, 2014). The RF model makes a prediction based on a set of hundreds of decision trees with identical distribution. The development of the trees may slow down the algorithm's prediction. Hyperparameters and tuning methods can be used to optimize RF. Tuning is the process of determining the best hyperparameters for a learning algorithm for a particular dataset. Aside from tuning, numerous ways for optimizing the RF model were suggested. These methods include grid search, F-race, the OOB approach and generic simulated annealing (Seibold *et al.*, 2017).

Heavy metal removal optimization

Heavy metal removal optimization by ZES varied depending on the zeolite dose, metal contents, water pH and temperature (Table 5). For Cu, optimized removal was achieved at 86.98% when the concentration of Cu in water was 5.685 mg/L. Optimization for Pb and Zn was different and was influenced more by pH and temperature than metal

Table 5: Heavy metal removal optimization parameters

Heavy metals	Removal efficiency (%)	Zeolite doce (g)	Metal contents (mg/L)	Water	
				pH	Temperature (°C)
Cu	86.98	0.08	5.685	4.69	29.5
Pb	98.92	0.08	28.035	4.67	29.0
Zn	95.97	0.074	1.575	5.2	29.9

contents in water. For Pb, the optimum pH and temperature were found to be <4.94 and 29.1 °C, respectively with 98.92% Pb reduction. In contrast, for 95.97% of Zn reduction, the optimum pH and temperature were found to be <5.61 and 29.7 °C.

CONTULATION

This study has shown that zeolite can be embedded in sheets by heating because the zeolite powder sticks to the sheet during the heating process. Additionally, the sandwich method used allows the zeolite powder embedded in the sheet to become denser. This study succeeded in showing the removal of heavy metals from water using ZES and modeling the contribution of variables that affect heavy metal removal using an RF model. The heavy metal removal efficiency by ZES was reduced to 50% at metal concentrations of 10.355 mg/L for Cu, 171.615 mg/L for Pb and 4.755 mg/L for Zn. According to the model, the important variables affecting the heavy metal removal efficiency of ZES were metal contents in water, followed by water temperature and pH. This modeling is supported by validation based on the RMSE, MSR, percentage variable explained and graphs depicting the OOB error. The RMSE values obtained in this study were in the order of Zn > Cu > Pb, whereas the MSR values obtained were in the order of Pb > Zn > Cu. On the basis of OOB error, all models show a large number of trees when the OOB error is decreasing, which indicates the model has good accuracy. Between heavy metals, the best model was observed for Zn and Cu, followed by that for Pb. An RF model can determine the threshold of heavy metal contents, water pH and temperature to optimize the heavy metal removal efficiency of ZES. Although this study was successful in predicting metal adsorption, the RF approach still has significant limitations that must be addressed. The RF model makes a prediction based on a set of hundreds of decision trees with identical distribution and the development of those trees may slow down the algorithm's prediction. In conclusion, high removal of Cu from water by ZES is observed if the Cu content is ≤5.685 mg/L. Conversely, the high removal of Pb and Zn from water by ZES is observed if the water pH ranges from 4.94 to 5.61 and the temperature is ≤29.1 °C. The application of this model can assist in the development of adsorbents in reducing pollutant levels in water. This study can also be a reference for environmentalists as an alternative material in handling waste that enters the water system.

AUTHOR CONTRIBUTIONS

N.D. Takarina as corresponding author has contributed in funding. N. Matsue provided references for the manuscript. E. Johan verified the data and results. A. Adiwibowo drafted the manuscript, analyzed and interpretated data using RF. M.F.N.K. Rahmawati conducted material preparation and fabrication of ZES. S.A. Pramudyawardhani helped in the Cu, Pb and Zn adsorption experiments using ZES. T. Wukirsari assisted in the preparation of stock solutions of Cu, Pb and Zn.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/or falsification, double publication and/or submission, and redundancy have been completely observed by the authors.

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ABBREVIATIONS

%	Percent
°C	Degree Celsius
δ	Symmetrical bending vibrations
/cm	Reciprocal centimeter
Al	Aluminum
Al–O	Aluminum–oxygen bonds
AlO_3	Aluminum oxide
ANN	Artificial neural networks
BPNN	Back propagation neural networks
BRIN	The National Research and Innovation Agency
C	Carbon
Ca	Calcium
C–H	Carbon–hydrogen
Cl	Chloride
CI	Confidence intervals
Cl	Chloride
cm^2	Centimeter squared
Cu	Copper
Cu^{2+}	Copper ion
$CuSO_4.5H_2O$	Copper (II) sulfate pentahydrate
Fe	Iron
F	Means between population / distribution
FTIR	Fourier transform infrared
g	Gram
GA	Genetic algorithms
H_i	Heavy metal concentration before treatment using ZES
H_t	Heavy metal concentration after treatment using ZES
i	A subset that contains the observations
ICP	Inductive couple plasma
ILRC	The Integrated Laboratory and Research Center University of Indonesia
L	Liter
LASSO	Least absolute shrinkage and selection operators
LASSO-GA-BPNN	Least absolute shrinkage and selection operators–genetic algorithms– back-propagation neural networks
LTA	Linde type A
Ltd	Limited
M	Molar
mg/L	Milligram per liter
mL	Milliliter
ML	Machine learning
mM	Millimolar
Mn	Manganese
$NaNO_3$	Sodium nitrate
Na_2SO_4	Sodium sulfate
MSR	Mean square of residuals
Na	Natrium
O	Oxygen
$O–CH_3$	Methoxy
O–Si–O	Siloxane – bending form
OOB	Out of bag error
OH	Oxygen hydrogen
P	Probability
pH	Potential of hydrogen
Pb	Lead
$Pb(NO3)_2$	Lead (II) nitrate
PE	Polyethylene
PP	Polypropylene

R^2	Coefficient of determinant
RF	Random forest
RMSE	Root mean square error
SEM-EDX	Scanning electron microscope–energy-dispersive X-ray
Si	Silicon
Si/Al	Silicon per aluminum
Si–O–Al	Silicon–oxygen–aluminum
Si–O–Si	Siloxane – stretching form
Si–O	Silicon–oxygen bonds
SiO_2	Silicon dioxide
SVR	Support vector regression
T	A real number which is defined by the algorithm
TiO_2	Titanium dioxide
V_{as}	Asymmetrical stretching vibrations
V_s	Symmetrical stretching vibrations
x_j	Variables
ZES	Zeolite-embedded sheet
Zn	Zinc
$ZnSO_4.7H_2O$	Zinc sulfate heptahydrate

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