



Performance evaluation of nanotubular halloysites from weathered pegmatites in removing heavy metals from water through novel artificial intelligence-based models and human-based optimization algorithm

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ARTICLE INFO

Handling Editor: X. Cao

Keywords:

Heavy metals
Water treatment
Nanotubular halloysites
Weathered pegmatites
Artificial intelligence
Optimization algorithm

ABSTRACT

The efforts of this study aimed to evaluate the feasibility of the nanotubular halloysites in weathered pegmatites (NaHWP) for removing heavy metals (i.e., Cd^{2+} , Pb^{2+}) from water. Furthermore, two novel intelligent models, such as teaching-learning-based optimization (TLBO)-artificial neural network (ANN), and TLBO-support vector regression (SVR), named as TLBO-ANN and TLBO-SVR models, respectively, were proposed to predict the Cd^{2+} and Pb^{2+} absorption efficiencies from water using the NaHWP adsorbent. Databases used, including 53 experiments for Pb^{2+} absorption and 56 experiments for Cd^{2+} absorption from water, under the catalysis of different conditions, such as initial concentration of Pb^{2+} and Cd^{2+} , solution pH, adsorbent weight, and contact time. Subsequently, the TLBO-ANN and TLBO-SVR models were developed and applied to predict the efficiencies of Cd^{2+} and Pb^{2+} absorption from water, aiming to evaluate the role as well as the effects of different conditions on the absorption efficiencies using the NaHWP adsorbent. The standalone ANN and SVM models were also taken into consideration and compared with the proposed hybrid models (i.e., TLBO-ANN and TLBO-SVR). The results showed that the NaHWP detected in a Kaolin mine (Vietnam) with 70% nanotubular halloysites is a potential adsorbent for water treatment to eliminate heavy metals from water. The two novel hybrid models proposed, i.e., TLBO-ANN and TLBO-SVR, also yielded the dominant performances and accuracies in predicting the Cd^{2+} and Pb^{2+} absorption efficiencies from water, i.e., $\text{RMSE} = 1.190$ and 1.102 , $\text{R}^2 = 0.951$ and 0.957 , $\text{VAF} = 94.436$ and 95.028 for the TLBO-ANN and TLBO-SVR models, respectively, in predicting the Pb^{2+} absorption efficiency from water; $\text{RMSE} = 3.084$ and 3.442 , $\text{R}^2 = 0.971$ and 0.965 , $\text{VAF} = 96.499$ and 96.415 for the TLBO-ANN and TLBO-SVR models, respectively, in predicting the Cd^{2+} absorption efficiency from water. Furthermore, the validation results also demonstrated these findings in practice through 23 experiments with the accuracies of 98.3% and 98.37% for the TLBO-ANN and TLBO-SVR models, respectively, in predicting the Pb^{2+} absorption efficiency from water; the accuracies of 98.3% and 97.46% for the TLBO-ANN and TLBO-SVR models, respectively, in predicting the Cd^{2+} absorption efficiency from water. Besides, solution pH was evaluated as the most critical parameter that can be adjusted to enhance the performance of the absorption of the heavy metals in this study. By using the NaHWP adsorbent and the novel proposed intelligent models developed, heavy metals can be eliminated entirely from water, providing pure water/clean freshwater without any risk of adverse health effects for the short term or long term.

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<https://doi.org/10.1016/j.chemosphere.2021.131012>

Received 10 April 2021; Received in revised form 21 May 2021; Accepted 24 May 2021

Available online 3 June 2021

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1. Introduction

Water is an indispensable part of human life, and it is one of the big concerns globally, especially in developing countries. According to the

Nomenclature

NaHWP	Nanotubular halloysites in weathered pegmatites
TLBO	Teaching-learning-based optimization
ANN	Artificial neural network
SVR	Support vector regression
AI	Artificial intelligence
RF	Random forest
KELM	Kernel extreme learning machine
SEM	Scanning electron microscopy
EDS	Energy-dispersive X-ray spectroscopy
TEM	Transmission electron microscopy
Al	Aluminum
Si	Silicon
O	Oxygen
Pb	Lead
Cd	Camidium
IP-MS	Plasma-mass spectrometric
RMSE	Root-mean-squared error
MSE	Mean squared error

World Bank estimation, about only 40% of the populations around the world have been used clean water resources for a specific end-use (Tortajada and Biswas, 2018). The remaining must use the raw water with many contaminations, such as heavy metals, bacteria, viruses, to name a few. Nowadays, due to rapid industrialization, many water supplies have been polluted (Rajput et al., 2017; Jayaswal et al., 2018; Cuc et al., 2020; Phong et al., 2020). Furthermore, due to rapid urbanization, the demands for water ingestion and water purification are increasing significantly. Many efforts have been adopted to survey and extract flowwater and groundwater (Binh et al., 2020; Dieu et al., 2020). However, they are still very scarce to provide for human life. Therefore, the clean freshwater problems (e.g., drinking water, water ingestion, and water purification) and how to treat raw water to achieve the desired end-use is the big concern of researchers worldwide.

Many techniques/technologies have been proposed to process/treat raw water/polluted water in recent years. The treatment is meaningful and crucial to human health, aiming to respond to the demands of clean freshwater without any risk of adverse health effects for the short term or long term. The techniques/technologies involve removing contaminants (i.e., heavy metals, harmful microbes, residual disinfectants) from raw water to produce cleaner water that is pure enough for human consumption (Bo et al., 2021a, 2021b). Of those, heavy metals are taken into account at a high-risk level to human health due to their high toxicity and carcinogenicity (Morais et al., 2012; Wallace and Djordjevic, 2020). To remove heavy metals, graphene oxide and its composites have been introduced as excellent and most potential adsorbents (Peng et al., 2017). Besides, carbon nanotubes have also been introduced as potential adsorbents for heavy metals removal (Stafiej and Pyrzynska, 2007; Tofighy and Mohammadi, 2011; Fiyadh et al., 2019). In addition, lignin and its derivatives are also proposed as the cost-effective adsorbent to absorb heavy metals in water (Wallace and Djordjevic, 2020). It is a fact that scientists' efforts in finding out new materials for water treatment are significant. In recent years, halloysite nanotubes have been discovered, and it was widely applied in various fields of industries (Danyliuk et al., 2020), especially for water treatment (Hebbar et al., 2016; Yu et al., 2016). It is a natural mineral and contained in soils and

rocks in some countries, such as China, the USA, France, Brazil, to name a few (Kausar, 2018). Despite the fact that halloysite nanotubes have been applied to remove heavy metals in water. Nevertheless, depending on each country's formation conditions, its properties are different (Pabakhsh et al., 2013). Therefore, this study introduced nanotubular halloysites in weathered pegmatites (NaHWP) – a type of halloysite discovered in an open-pit kaolin mine in Vietnam with 70% of halloysite included in the materials, for the removal of heavy metals. Herein, NaHWP was considered to remove Pb^{2+} and Cd^{2+} from drinking water.

In water treatment, measures and predictions of treatment efficiency also play an essential role in processes. Measures are taken to ensure the quality of water after treatment. In some cases, water quality is evaluated through its distribution and conveyance after treatment (Delahaye et al., 2003; Shrestha et al., 2011; Rezaei et al., 2019). During the absorption of heavy metals by various adsorbents, the absorption efficiency of different adsorbents and different conditions is not the same. Thus, the prediction of absorption efficiency is necessary to understand the properties of the adsorbent used, as well as the necessary conditions to achieve the highest performance. In recent years, artificial intelligence (AI) and machine learning methods have been widely applied in real-life with promising results (Hoang et al., 2020). Indeed, several researchers applied different AI techniques to forecast the efficiency of heavy metals absorption from water. Zhu et al. (2019) applied artificial neural network (ANN) and random forest (RF) models for predicting the absorption efficiency of heavy metals on biochar. They found that the RF model provided a promising performance for this aim with R^2 of 0.973. In another study, Zhao et al. (2021) applied the kernel extreme learning machine (KELM) and Kriging models for similar purposes. They used these models for not only the prediction of the absorption efficiency of heavy metals but also for control factors on metal absorption. By the use of another machine learning algorithm, such as support vector machine (SVR), Li et al. (2019) also successfully predicted the absorption efficiency of Pb^{2+} from water on biochar. The accuracy of 100% was reported in their study for the biochar system used and the efficiency of Pb^{2+} absorption. Similar studies with similar AI models have also been conducted, and the readers can refer to the literature (Ahmad et al., 2018; Moreno-Pérez et al., 2018; Lata et al., 2019). The literature review indicates that several AI methods have been proposed to predict heavy metals' absorption efficiency from water. Nevertheless, they have not yet applied to all materials.

Moreover, novel methods/models with the accuracy improved are the goal of researchers to achieve the highest performance in this term. Therefore, in this study, two novel AI models based on the hybridization of teaching-learning-based optimization (TLBO)- ANN, and TLBO- SVR, named TLBO-ANN and TLBO-SVR models, were proposed to predict the absorption efficiency of Pb^{2+} and Cd^{2+} from water using the NaHWP material, as described earlier. These models aim to provide insights into the absorption efficiency of heavy metals (i.e., Pb^{2+} and Cd^{2+}) by the NaHWP. Also, the input factors, as well as the absorption conditions, can be controlled and optimized by these models to maximum eliminate heavy metals from drinking water using NaHWP. The structure of this paper is organized as follows:

- Section 1: Introduction of the research problem and review of the related works.
- Section 2: Introduction of the study site and general geological structure.
- Section 3: Introduction of the nanotubular halloysites in weathered pegmatites detected in the study area.
- Section 4: Experiments of heavy metals absorption and the dataset used for this study.
- Section 5: Proposing the framework of the novel hybrid intelligent models for predicting the heavy metals absorption efficiency.
- Section 6: Development of artificial intelligent-based models for predicting the heavy metals absorption efficiency.
- Section 7: Presentation of the obtained results.

- Section 8: Discussion of the obtained results.
- Section 9: Validation of the novel intelligent models developed.
- Section 10: Sensitivity analysis to highlight the importance of the input variables used.
- Section 11: Conclusion and remarks.

2. Study site and general geological structure

The study area (Thach Khoan, Phu Tho province) is about 85 km northwest of Hanoi city, the capital of Vietnam. Most of the study area is occupied by the metamorphic Thach Khoan Formation of Proterozoic age (PR₃-Є₁tk). Next are the early Devonian Ban Nguon Formation (D1bn) sedimentary rocks and loose Quaternary. Magma blocks related to the Late Paleozoic Tan Phuong granite complex are scattered distributed in the area. Remarkably, there are many pegmatite bodies with the strikes of N60° to 80°W and a dip of 50° ÷ 80° to the southwest in the area. The length of these pegmatite bodies ranges from a few hundred to thousands of meters, and these wide are from ten to hundreds of meters. (Fig. S1 in supplementary material). The pegmatite bodies are weathered, and the weathering profile is up to 45 ÷ 50 m. Tabular halloysites were identified in this weathered zone with significant amounts (Bui and Nguyen, 2016; Bac et al., 2018).

3. Nanotubular halloysites in weathered pegmatites

The sample collecting from the Thach Khoan area (Phu Tho province, Vietnam) was well mixed and then separated to the particle size of <32 μm using a wet sieving method. The sample under the sieve (<32 μm) was dried at 60 °C (Fig. 1A). Analytical methods used to determine the existence of tubular halloysites included scanning electron microscopy (SEM) with Fourier-transform infrared spectroscopy (FT-IR), energy-dispersive X-ray spectroscopy (EDS), and transmission electron microscopy (TEM).

Fig. 1B presents the sample's SEM image with halloysite minerals in rod-shaped shapes overlapping to a matrix. The EDS result for the mineral also indicates the presence of aluminum (Al), silicon (Si), and oxygen (O) elements, corresponding to the general chemical formula of the kaolin group (Al₂Si₂O₅(OH)₄.nH₂O) (Fig. 1C). The FT-IR analysis result is shown in Fig. 1D. From the FT-IR spectra, the vibration of stretching was defined with the absorptions bands at 3696 and 3620 cm⁻¹ since the O-H groups inner-surface; the interlayer water was defined with the bands at 1641 cm⁻¹; the stretching region of Si-O was defined with two bands at 1112 and 1006 cm⁻¹; the Al-OH bending vibration was defined with the band at 910 cm⁻¹; the Al-O-OH stretching mode was observed with the band at 796, 755 cm⁻¹ for the whole samples; the Si-O low stretching was defined with a band of 691 cm⁻¹; Al-O-Si and Si-O-Si vibration were defined with the bands at 540 and 471 cm⁻¹, respectively. The FT-IR pattern of this sample is similar to previous studies about halloysite minerals (Iuliu et al., 2001; Ray et al., 2000; Li et al., 2019). Notably, the TEM image clearly shows the tubular structure of the halloysite minerals in the sample. (Fig. 1E). Thus, the analysis results using SEM-EDS and TEM confirmed the existence of halloysite minerals in the Thach Khoan sample, consistent with previous research (Bui and Nguyen, 2016; Bac et al., 2018).

4. Experiments of heavy metals absorption and the dataset used for intelligent methods

The experiments are conducted in the laboratory by adding a quantity of nanotubular halloysite powder to 50 ml of a solution for Pb²⁺ (or Cd²⁺). The primary purpose of this task is to examine the effects of different physicochemical characteristics on absorption. The initial concentrations of Pb²⁺ (or Cd²⁺) are prepared and controlled in the range of from 20 ÷ 80 mg/L. The contact time for both ions is varied from 10 to 120 min. The solutions' pH is adjusted in a range of 2.95–8.8 (Supplementary material). The dose of nanotubular halloysite powder

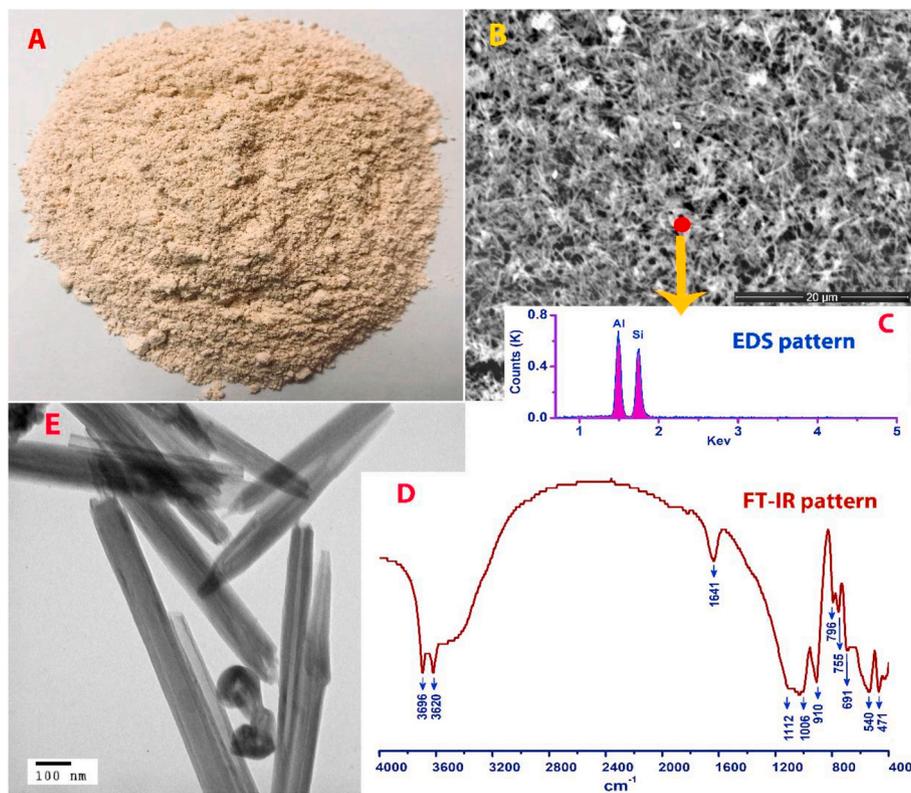


Fig. 1. Samples and tubular structure of the halloysite minerals. A - Sample of the halloysite minerals; B - Sample's SEM image with halloysite minerals in rod-shaped shapes overlapping to a matrix; C - Presence of elements in halloysite mineral through the EDS pattern; D - The FT-IR analysis results; E - TEM image of the halloysite minerals with the tubular structure.

changed from 0.3 to 0.9 g to obtain the unsaturated states. The mixtures are then shaken continuously at 100 rpm using a mechanical shaker at room temperature. Once the solid was removed by the filtration method, the remaining concentration of Pb^{2+} and Cd^{2+} was determined by using the inductively inducing plasma-mass spectrometric method (ICP-MS).

Finally, a total of 53 experimental cases were performed, with each case comprising five factors of the initial concentration of Pb^{2+} ($Pb_{initial}$), the adsorbent weight (Pb_{AW}), solution pH (Pb_{pH}), contact time (Pb_{CT}), and the Pb^{2+} concentration after absorption (Pb_{output}). For Cd^{2+} ion, 56 experimental cases were carried out, and five experimental factors are $Cd_{initial}$, Cd_{pH} , Cd_{AW} , Cd_{CT} , and Cd_{output} .

The details and characteristics of the dataset used are illustrated in Fig. S2 (Supplementary material), and the datasets used are also available in the Supplementary material with Tables S1 and S2.

5. Hybrid TLBO-ANN and TLBO-SVR models

As introduced above, ANN and SVR are robust and conventional soft computing models, and they have been widely applied in various areas (Chahnasir et al., 2018; Shariati et al., 2019, 2020; Hoang et al., 2021a, 2021b; Hong et al., 2021; Jue et al., 2021; Libing et al., 2021; Xuan-Nam et al., 2021). In this study, ANN and SVR were selected as the primary models for predicting the absorption efficiency of heavy metals from water (i.e., Pb^{2+} and Cd^{2+}). Based on the advantages and the robustness of the TLBO algorithm, the ANN and SVR models were optimized by the TLBO algorithm to improve their accuracy in predicting the absorption efficiency of Pb^{2+} and Cd^{2+} from water, named as TLBO-ANN and TLBO-SVR models. To optimize the ANN model, the TLBO algorithm implements optimization solutions to optimize the weights of the ANN model to find out their best values with the best performance of the ANN model. Whereas, the searching procedure of the TLBO algorithm implements a random search based on its optimization solutions to determine the optimal values of the SVR's hyper-parameters (e.g., σ and C). The principle of the ANN, SVR, and TLBO was introduced in the Supplementary material. The framework of the TLBO-ANN and TLBO-SVR models is proposed in Fig. 2.

Accordingly, the dataset was divided into two sections: training and testing. Subsequently, the training dataset was pre-processed and normalized in the range of [-1,1] before developing the models aiming to avoid overfitting of the models (Hoang, 2020). An ANN and SVR models were then developed based on the processed training dataset as the initial models. Whereas weights are the main parameter that can be used to control and tune the performance/accuracy of the ANN model, the SVR model used σ and C parameters to control and tune the

performance/accuracy of the model with the support of RBF function. Subsequently, the TLBO algorithm was used to optimize these parameters (i.e., weights, σ and C), aiming to provide better models with the performance/accuracy improved. Herein, root-mean-squared error (RMSE) was used as the objective function to optimize the ANN and SVR models. The stopping condition was then checked whether it is satisfied or not. Finally, the optimal TLBO-ANN and TLBO-SVR models were defined, and their performance is validated through the testing dataset.

6. Development of artificial intelligent-based models

Prior to developing the predictive models, the datasets are divided into 70% for the training and developing models and the remaining 30% for testing the models (Trung et al., 2019). The MinMax scaling method was applied with the range of [-1, 1] to avoid overfitting, as mentioned above.

6.1. ANN model

As is known, the network topology of an ANN model is a challenging before training the network for the aim of Pb^{2+} and Cd^{2+} absorption efficiency prediction. Therefore, a procedure of trial-and-error was implemented to define the structure of the ANN models (Mohammadhassani et al., 2013). Finally, an ANN topology with one hidden layer and 12 hidden neurons was defined for this stage. It is worth mentioning that the backpropagation algorithm was used to train the ANN model for estimating the efficiency of Cd^{2+} and Pb^{2+} absorption from water. Subsequently, deep learning techniques, such as the use of the "ReLU" active function, "he_uniform" kernel initializer, stochastic gradient descent optimizer with a learning rate of 0.1, the momentum of 0.9 and decay of 0.0001, were applied to train the ANN models, aiming to achieve the highest performances. Mean squared error (MSE) was used as the loss function to evaluate the performance during the development of the ANN models. The models' development results are shown in Fig. S6 (Supplementary material).

The performance curves in Fig. S6 indicated that the ANN models for predicting Pb^{2+} and Cd^{2+} absorption efficiency from water were well-developed without overfitting. In other words, these ANN models are good, and their performances are discussed and evaluated in the next section.

6.2. SVR model

Regarding the SVR modeling, the RBF function was applied to train

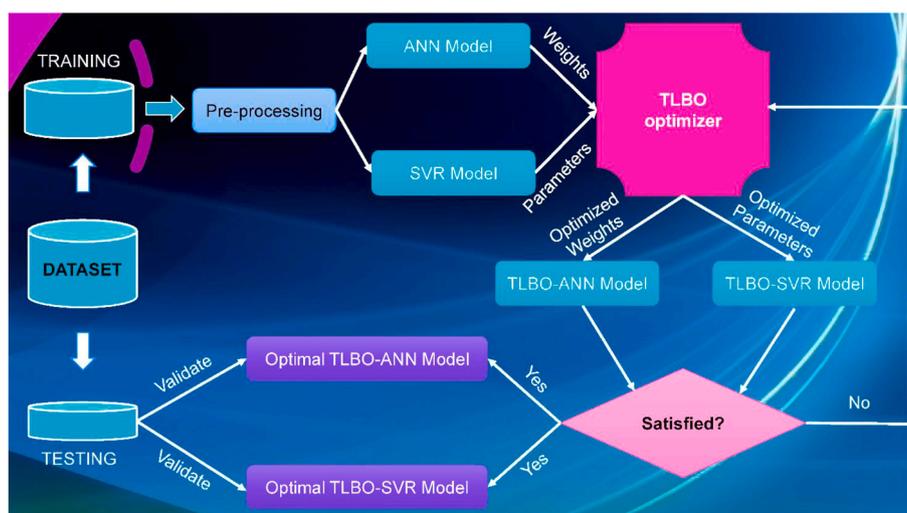


Fig. 2. The proposed framework of the TLBO-ANN and TLBO-SVR models for predicting heavy metals absorption in this study.

the SVR models for predicting Pb^{2+} and Cd^{2+} absorption efficiency from water. Accordingly, σ and C were used as the main parameters to tune the performance of the SVR models in this case. Grid search technique with 980 models was implemented to find out the best SVR models for predicting Pb^{2+} and Cd^{2+} absorption efficiency from water. Finally, the best SVR models were found with $\sigma = 0.1$ and $C = 10$ for this aim, and their performances are discussed and evaluated in the next section.

6.3. TLBO-ANN and TLBO-SVR models

Once the best ANN and SVR models were well-developed as the standalone models for predicting Pb^{2+} and Cd^{2+} absorption efficiency from water, the proposed framework in Fig. 2 was applied to develop the TLBO-ANN and TLBO-SVR models for similar purposes.

Accordingly, the parameters of the developed ANN and SVR models in the previous sections (i.e., weights, σ and C) were optimized by the TLBO algorithm, aiming to improve their performance/accuracy. To do this, the TLBO algorithm was set up with the number of learners are 50, 100, 150, 200, 250, 300, 350, 400, 450, 500, respectively. RMSE was used as the loss function to evaluate the optimization processes. The global search of the TLBO algorithm was repeated with 1000 iterations to determine the best position with the best learner. Finally, the optimal TLBO-ANN and TLBO-SVR models were defined through the training curves in Fig. 3. Their results and performances are shown, discussed, and evaluated in the next sections.

7. Results

As stated above, the testing datasets were used to validate the developed models (i.e., ANN, SVR, TLBO-ANN, and TLBO-SVR). The prediction results of the individual models on the Pb^{2+} and Cd^{2+} testing datasets are shown in Tables 1 and 2, respectively. Finally, their performances are computed in Table 3.

Table 1

Prediction results of the individual models for predicting Pb^{2+} absorption efficiency from water (testing phase).

Actual Pb^{2+} absorption efficiency	Predicted Pb^{2+} absorption efficiency			
	ANN	SVR	TLBO-ANN	TLBO-SVR
4.820	5.319	6.824	5.887	6.506
3.066	5.348	5.657	2.499	3.507
2.500	3.061	8.707	4.669	5.024
14.520	15.118	14.772	14.717	14.367
10.420	9.602	8.138	9.230	9.259
14.110	12.288	11.963	13.175	12.074
4.000	5.228	4.898	4.548	4.064
13.530	11.453	11.999	13.240	13.356
4.664	3.828	5.799	5.243	5.484
8.210	7.434	6.965	7.605	7.956
3.990	2.286	5.523	2.919	2.635
15.370	16.922	14.559	15.048	14.466
14.774	16.518	12.619	16.919	13.878
15.587	16.618	14.898	17.962	15.709
11.200	10.572	10.455	11.952	11.488
4.015	5.973	5.249	3.008	4.446

8. Discussion

In this section, two main objectives of this study are discussed and evaluated as mentioned in the title of the paper, including:

- (i) The feasibility of nanotube-type halloysite in weathered pegmatites for heavy metals removal (i.e., Pb^{2+} and Cd^{2+}) from water;
- (ii) The performances of the developed state-of-the-art models for predicting Pb^{2+} and Cd^{2+} absorption efficiency from water using nanotube-type halloysite in weathered pegmatites.

It can be seen that the modified nanotubular halloysites from

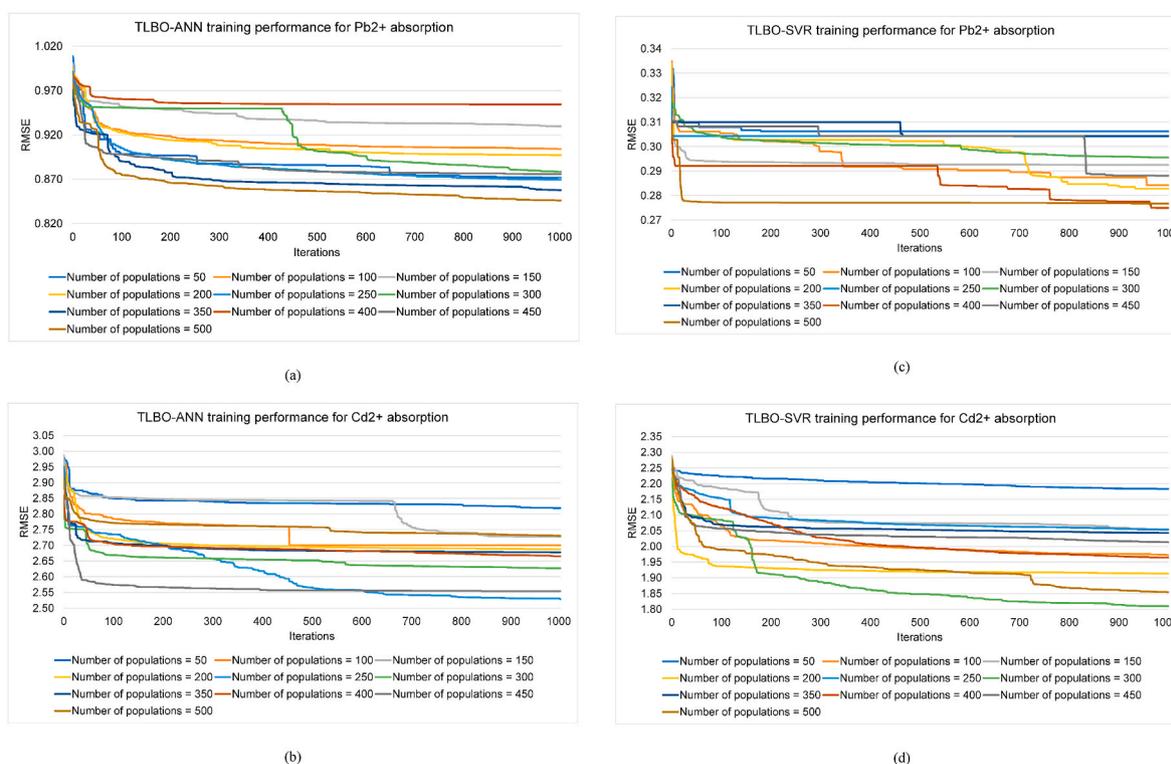


Fig. 3. Training performance of the TLBO-ANN and TLBO-SVR models during predicting Pb^{2+} and Cd^{2+} absorption efficiency from water (a) TLBO-ANN model for predicting Pb^{2+} absorption efficiency from water; (b) TLBO-ANN model for predicting Cd^{2+} absorption efficiency from water; (c) TLBO-SVR model for predicting Pb^{2+} absorption efficiency from water; (d) TLBO-SVR model for predicting Cd^{2+} absorption efficiency from water.

Table 2
Prediction results of the individual models for predicting Cd²⁺ absorption efficiency from water (testing phase).

Actual Cd ²⁺ absorption efficiency	Predicted Cd ²⁺ absorption efficiency			
	ANN	SVR	TLBO-ANN	TLBO-SVR
22.087	20.837	17.489	17.658	20.213
40.617	43.779	38.166	44.190	37.849
41.667	43.441	37.488	42.715	38.119
34.114	30.941	39.055	33.204	35.028
18.076	19.791	15.597	19.189	20.258
17.852	19.319	18.121	19.543	20.932
17.603	20.061	23.078	18.874	18.075
59.261	63.280	53.553	61.013	58.804
22.952	21.282	22.341	21.785	22.247
60.619	66.779	50.506	63.615	55.126
32.636	27.125	31.294	32.404	30.632
19.299	17.249	23.351	20.472	20.463
58.344	58.290	55.118	59.921	60.218
23.521	29.261	16.905	29.441	14.781
16.431	14.164	19.485	13.816	14.524
39.256	29.662	38.897	32.194	36.587
60.890	68.274	51.224	64.431	54.933

weathered pegmatites have a good ability to adsorb Pb and Cd ions in water. The absorption efficiency of this material is higher than the flat kaolinite, which is also one of the minerals of the kaolin group (Angove et al., 1997; Srivastava et al., 2004; Maged et al., 2020). This difference may be due to a certain effect of small particle sizes and the unique tubular structure of the halloysites. Additionally, halloysites have multiple surface groups with aluminol groups (Al-OH) inside the tubes and siloxanes on the surfaces of tubes, which can form a charged surface for ion adsorption and depend on pH changes. The adsorption capacity of heavy metal ions in an aqueous solution can be increased when the halloysites in this area are modified under suitable experimental conditions.

Regarding the performances of the developed state-of-the-art models for predicting Pb²⁺ and Cd²⁺ absorption efficiency from water using nanotube-type halloysite in weathered pegmatites, as computed and listed in Tables 1–3, it can be seen that all four models are good in terms of heavy metals absorption (e.g., Pb²⁺ and Cd²⁺). Notably, the hybrid models, i.e., TLBO-ANN and TLBO-SVR, provided better performances than those of the standalone ANN and SVR models. In other words, the TLBO algorithm improved the accuracy of the standalone ANN and SVR models significantly. In this sense, the accuracy of the TLBO-ANN model was improved by approximately 2.2% in predicting Pb²⁺ absorption efficiency and improved approximately 3% in predicting Cd²⁺ absorption efficiency compared with the standalone ANN model. Whereas, the TLBO-SVR model was improved by approximately 14% in predicting Pb²⁺ absorption efficiency and improved approximately 4.3% in predicting Cd²⁺ absorption efficiency compared with the standalone SVR model. Figs. 4 and 5 show the correlation and box-whisker plots of the models, for both training and testing dataset, in predicting Pb²⁺ and Cd²⁺ absorption efficiency, respectively.

Table 3
Performances of the individual models for predicting Pb²⁺ and Cd²⁺ absorption efficiency from water.

Heavy metals absorption	Models	Training phase			Testing phase		
		RMSE	R ²	VAF	RMSE	R ²	VAF
Pb ²⁺ absorption	ANN	1.320	0.961	96.047	1.389	0.924	92.217
	SVR	1.923	0.948	91.626	2.170	0.849	81.001
	TLBO-ANN	0.846	0.984	98.375	1.190	0.951	94.436
	TLBO-SVR	0.275	0.999	99.830	1.102	0.957	95.028
Cd ²⁺ absorption	ANN	3.089	0.952	95.248	4.284	0.951	93.115
	SVR	4.227	0.919	91.156	4.945	0.938	92.187
	TLBO-ANN	2.528	0.968	96.817	3.084	0.971	96.499
	TLBO-SVR	1.810	0.985	98.425	3.442	0.965	96.415

For the Pb²⁺ absorption efficiency predictive models, the predictions' trend is good, as shown in Fig. 4; however, their convergence in the regression lines is limited, especially the convergence of the SVR predictions. Furthermore, the SVR model provided some outcomes out of the model's 80% confidence level. Meanwhile, the number of such outcomes is less for the ANN model. Considering the box plots of the models in predicting Pb²⁺ absorption, it can be seen that the distribution, as well as accuracy of the developed models, are in good forms, especially the TLBO-ANN model. No outlier appeared in the TLBO-ANN, TLBO-SVR, and ANN models; whereas, only one outlier appeared in the training phase of the SVR model.

Regarding the Cd²⁺ absorption efficiency predictive models (Fig. 5), the results seem to be better than the Pb²⁺ absorption efficiency predictive models, and most of the predictions are fit in the 80% of confidence level. Indeed, the accuracy of the AI models is highly appreciated in predicting Cd²⁺ absorption efficiency, especially the TLBO-ANN model. To have a comprehensive assessment and view of the developed models' accuracy in practical, the Taylor diagrams were calculated and visualized in Fig. 6.

Looking at Fig. 6 can see that the accuracy of the hybrid models, i.e., TLBO-ANN and TLBO-SVR, are higher than those of the ANN and SVR model without optimization of the TLBO algorithm. Whereas the accuracy of the TLBO-SVR model is slightly higher than the TLBO-ANN model in predicting Pb²⁺ absorption efficiency, their performance is similar in predicting Cd²⁺ absorption efficiency, and both are close to the observed model (actual model).

9. Validation of the TLBO-ANN and TLBO-SVR in practice

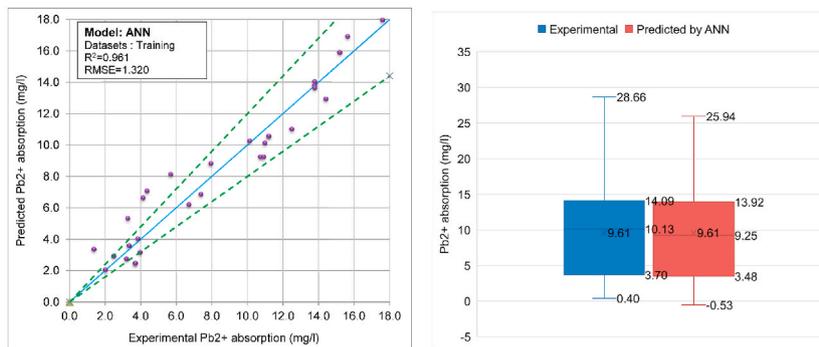
Due to the TLBO-ANN and TLBO-SVR models were defined as the best models for predicting the efficiency of Cd²⁺ and Pb²⁺ absorption from water; therefore, they were selected to validate in practice. Accordingly, we have performed further experiments, including 11 samples for Pb²⁺ absorption and 12 samples for Cd²⁺ absorption under various conditions of the inputs, as listed in Tables 4 and 5.

Once the validation datasets of Cd²⁺ and Pb²⁺ absorption from water were prepared, the TLBO-ANN and TLBO-SVR models were applied to predict the efficient absorption of Cd²⁺ and Pb²⁺ from water. The prediction results are illustrated in Fig. 7, and their performance is computed in Table 6.

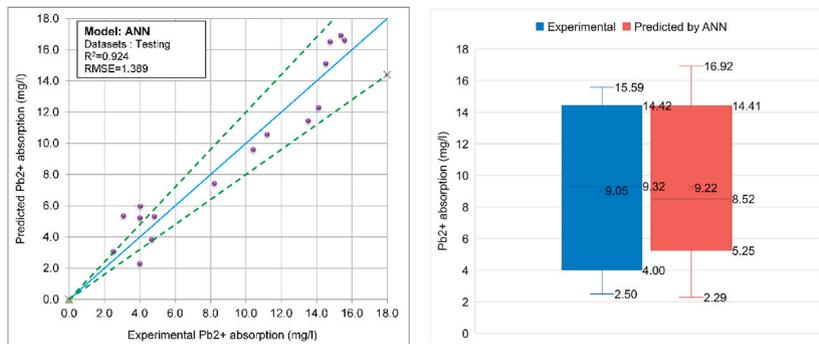
From the obtained validation results based on the further experiments, it can be seen that NaHWP is a sturdy material to absorb heavy metals from water, especially Pb²⁺ and Cd²⁺. Furthermore, the TLBO-ANN and TLBO-SVR models are robust and high reliable models for predicting the efficiency of Cd²⁺ and Pb²⁺ absorption from water in practice.

10. Sensitivity analysis

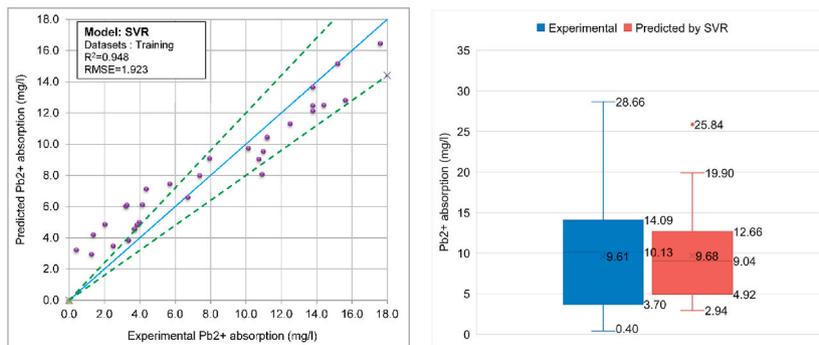
After the NaHWP and the TLBO-ANN and TLBO-SVR models were well-evaluated and defined for removing heavy metals (e.g., Pb²⁺ and Cd²⁺) in water treatment under various inputs and conditions, a sensitivity analysis was conducted to determine the importance of the input



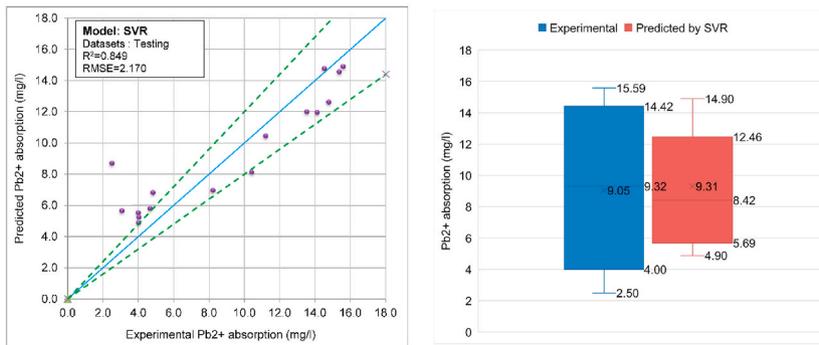
(a)



(b)

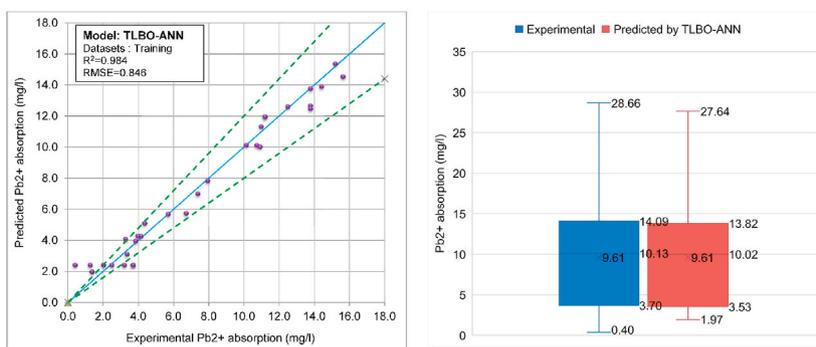


(c)

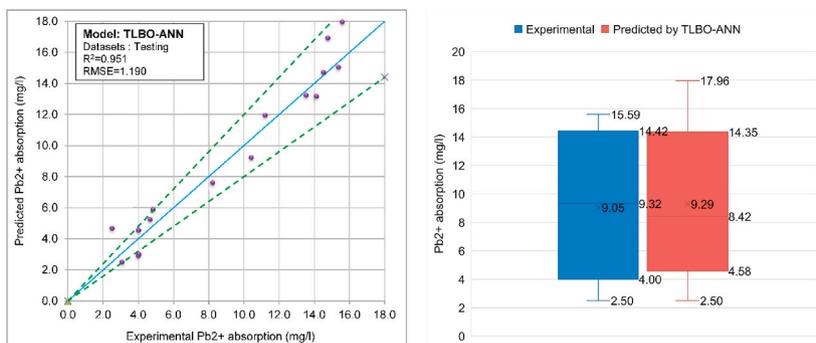


(d)

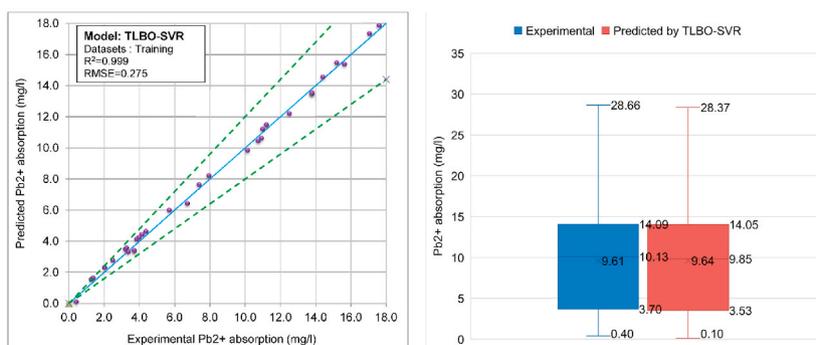
Fig. 4. Training and testing accuracies of the individual models in predicting Pb^{2+} absorption efficiency from water through correlation and box plots. (a) Training ANN model; (b) Testing ANN model; (c) Training SVR model; (d) Testing SVR model; (e) Training TLBO-ANN model; (f) Testing TLBO-ANN model; (g) Training TLBO-SVR model; (h) Testing TLBO-SVR model.



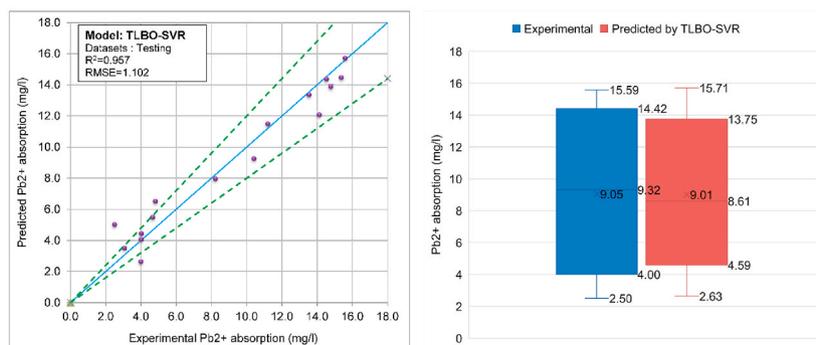
(e)



(f)



(g)



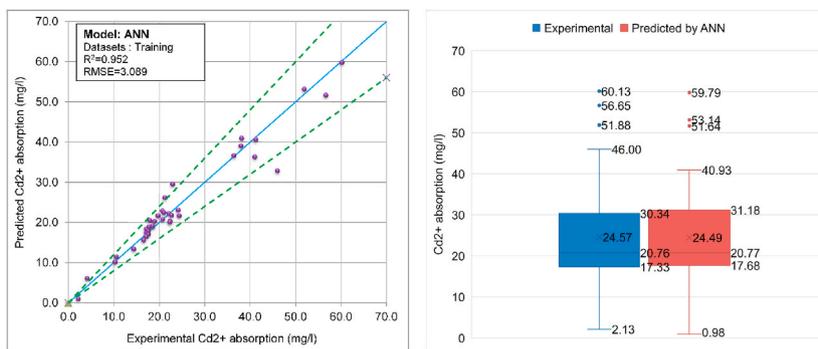
(h)

Fig. 4. (continued).

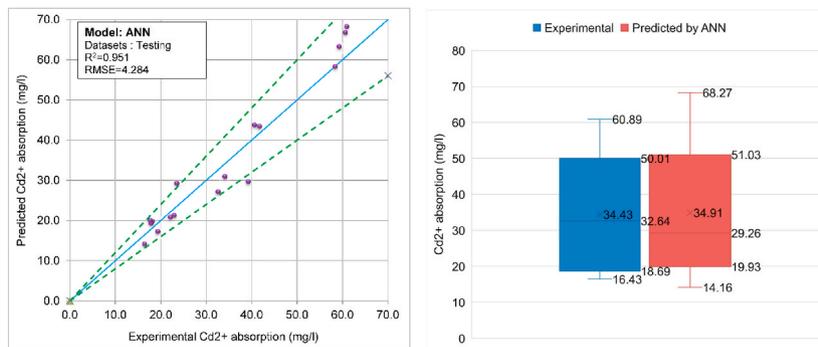
variables, as shown in Fig. S7 (Supplementary material). This task aims at providing a comprehensive solution to adjust the values of the input variables aiming to achieve higher performance in removing heavy

metals from water using NaHWP and the developed AI models.

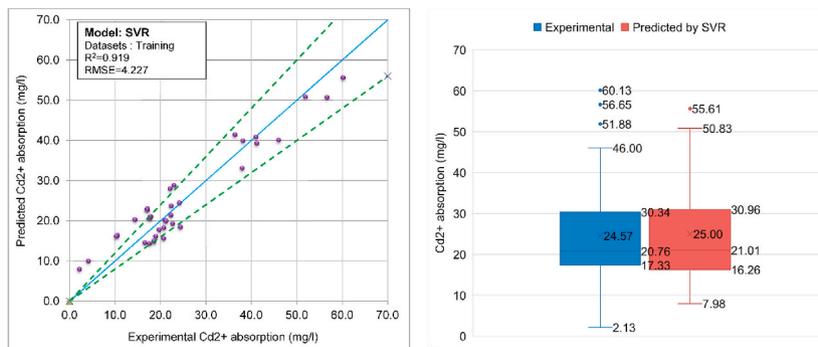
Based on Fig. S7, it can be seen that the initial concentrations of Pb^{2+} and Cd^{2+} are the most important, following are the pH, AW and CT.



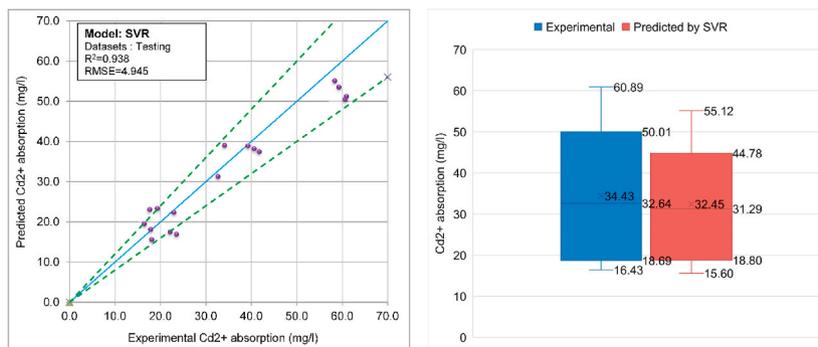
(a)



(b)

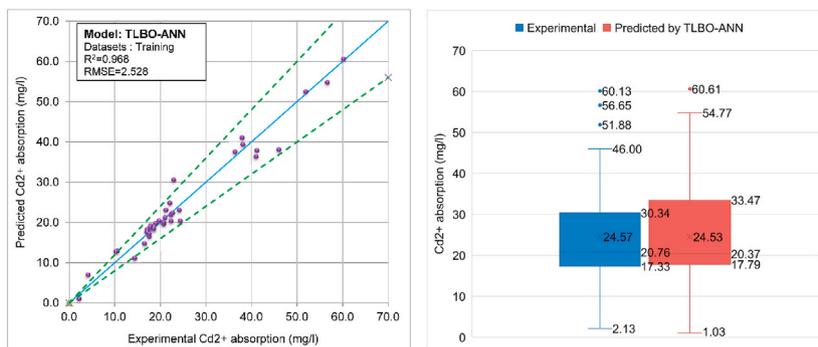


(c)

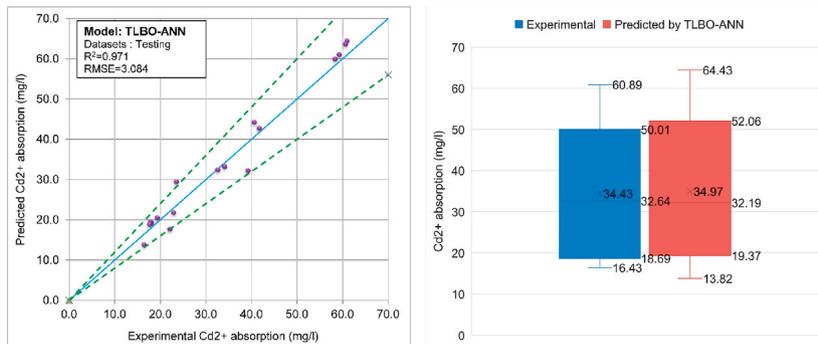


(d)

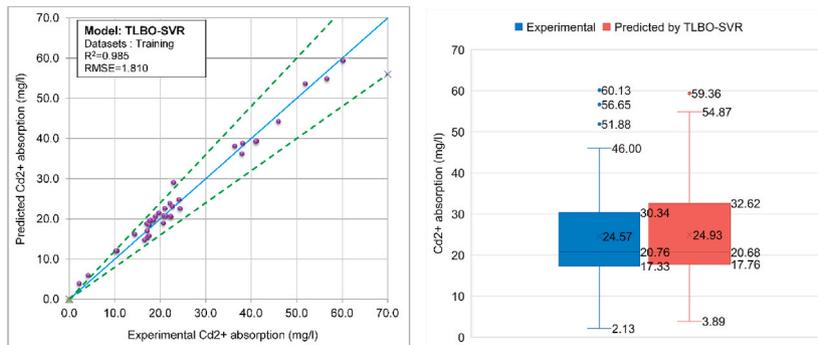
Fig. 5. Training and testing accuracies of the individual models in predicting Cd²⁺ absorption efficiency from water through correlation and box plots (a) Training ANN model; (b) Testing ANN model; (c) Training SVR model; (d) Testing SVR model; (e) Training TLBO-ANN model; (f) Testing TLBO-ANN model; (g) Training TLBO-SVR model; (h) Testing TLBO-SVR model.



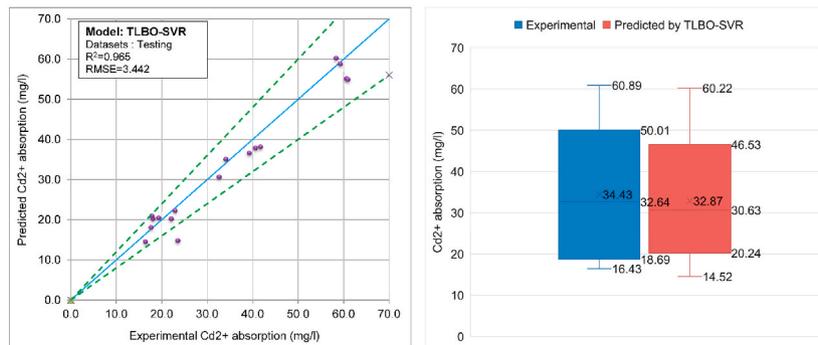
(e)



(f)



(g)

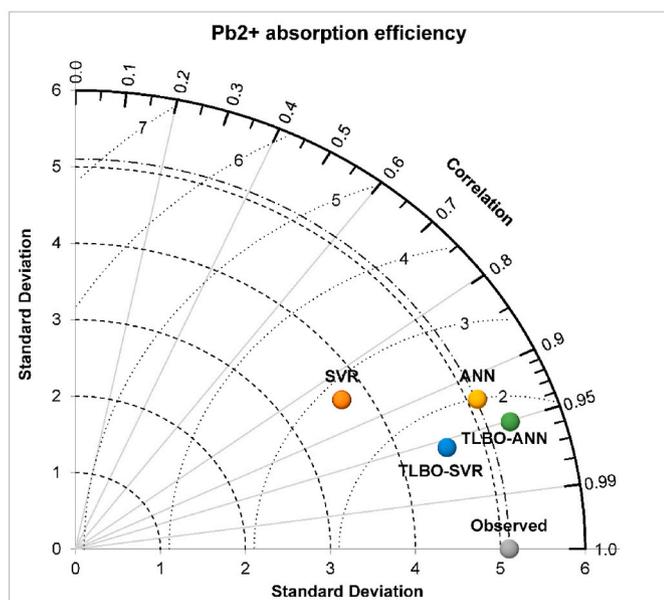


(h)

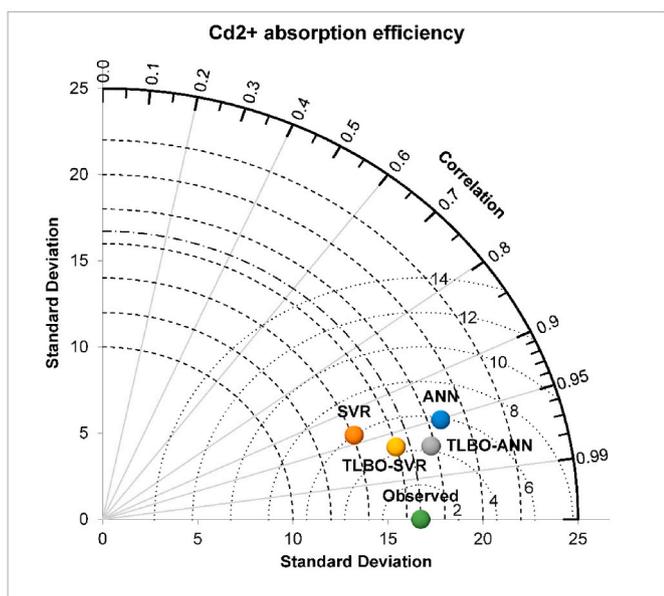
Fig. 5. (continued).

Among the four inputs used, CT appears to have a minor effect on heavy metal removal performance in water using NaHWP for both Pb^{2+} and Cd^{2+} . Whereas the $Pb_{initial}$ and $Cd_{initial}$ are uncontrollable parameters,

pH can be adjusted to enhance the performance of the absorption of the heavy metals in this study.



(a)



(b)

Fig. 6. Taylor diagrams of the individual models for evaluating the prediction efficiency (a) Pb^{2+} absorption; (b) Cd^{2+} absorption.

Table 4
Validation dataset of Pb^{2+} absorption from water.

$Pb_{initial}$	Pb_{pH}	Pb_{AW}	Pb_{CT}	Pb_{output}
35	4.96	0.6	60	6.102
45	4.96	0.6	60	12.022
45	4.96	0.6	65	12.125
40	5.31	0.5	35	4.289
55	4.96	0.4	60	38.127
55	4.96	0.4	65	38.029
40	5.31	0.6	50	2.223
20	5.31	0.7	60	2.015
80	5.31	0.6	60	2.425
60	5.31	0.7	55	3.178
70	5.31	0.7	60	4.369

Table 5
Validation dataset of Cd^{2+} absorption from water.

$Cd_{initial}$	Cd_{pH}	Cd_{AW}	Cd_{CT}	Cd_{output}
80	6.3	0.4	60	60.143
80	6.3	0.5	65	44.012
70	6.8	0.5	60	40.759
70	6.8	0.6	75	31.854
46	6.2	0.5	55	23.669
46	6.2	0.6	65	23.864
50	6.5	0.8	60	15.045
60	6.3	0.8	70	25.369
70	6.2	0.7	65	43.992
80	6.5	0.6	50	61.859
20	6.5	0.7	65	4.857
30	6.5	0.8	60	3.567

11. Conclusion, remarks and limitations

Water treatment with the removal of heavy metals is meaningful and crucial to human health, aiming to respond to clean freshwater demands without any risk of adverse health effects for the short term or long term. In the theme of this work, we discovered the nanotubular halloysites from weathered pegmatites in a kaolin mine with 70% of nanotubular halloysites as a potential material for absorption of heavy metals (i.e., Pb^{2+} and Cd^{2+}) from water. Their feasibility was also thoroughly evaluated in terms of removing heavy metals (i.e., Pb^{2+} and Cd^{2+}) by the chemical-physical method. The experiments and results indicated that this material is a good absorbent for removing Pb^{2+} and Cd^{2+} from water, with 79.3% of Pb^{2+} were removed and 51.45% of Cd^{2+} was removed from the water.

Although the performance of the heavy metals' absorption from the water of this material is good, efforts to improve the performance of the absorption of the heavy metals from the water of this material are necessary. The experiments show that the performance of the absorption of the heavy metals from water depends on the initial concentration of Pb^{2+} or Cd^{2+} , solution pH, adsorbent weight, and contact time. Therefore, we developed and proposed two novel hybrid intelligent models for predicting the heavy metals absorption efficiency from water based on these parameters, namely TLBO-ANN and TLBO-SVR models. The results show the proposed TLBO-ANN and TLBO-SVR models can predict the heavy metals absorption efficiency from water with high accuracy. In other words, they can be used to adjust the input parameters to achieve higher performance in removing heavy metals from water.

Inspire of the absorption performance of the discovered material as well as the accuracy of the proposed intelligent models (i.e., TLBO-ANN and TLBO-SVR) are outstanding in this study; nevertheless, some limitations are necessary for further research in the future:

- The feasibility of this material in the absorption of other toxic heavy metals from water, such as Hg^{2+} , Cr^{4+} , Ni^{2+} , Cu^{2+} , to name a few, should be considered and evaluated in future research.
- A larger dataset of heavy metals absorption is better for the proposed intelligent models aiming to provide higher accuracies in predicting the heavy metals absorption efficiency from water. With the higher accuracies of the models, heavy metals in water can be removed in a better way.

Credit author statement

Bui Hoang Bac and Hoang Nguyen: Conceptualization, Methodology, Software, Visualization, Writing and preparing the original version and revised version of the manuscript. Vo Thi Hanh, Le Thi Duyen: Data analyze, Data curation. Nguyen Tien Dung, Nguyen Thi Thanh Thao, Luong Quang Khang, Do Manh An: Conceptualization, Methodology, Software, Visualization.

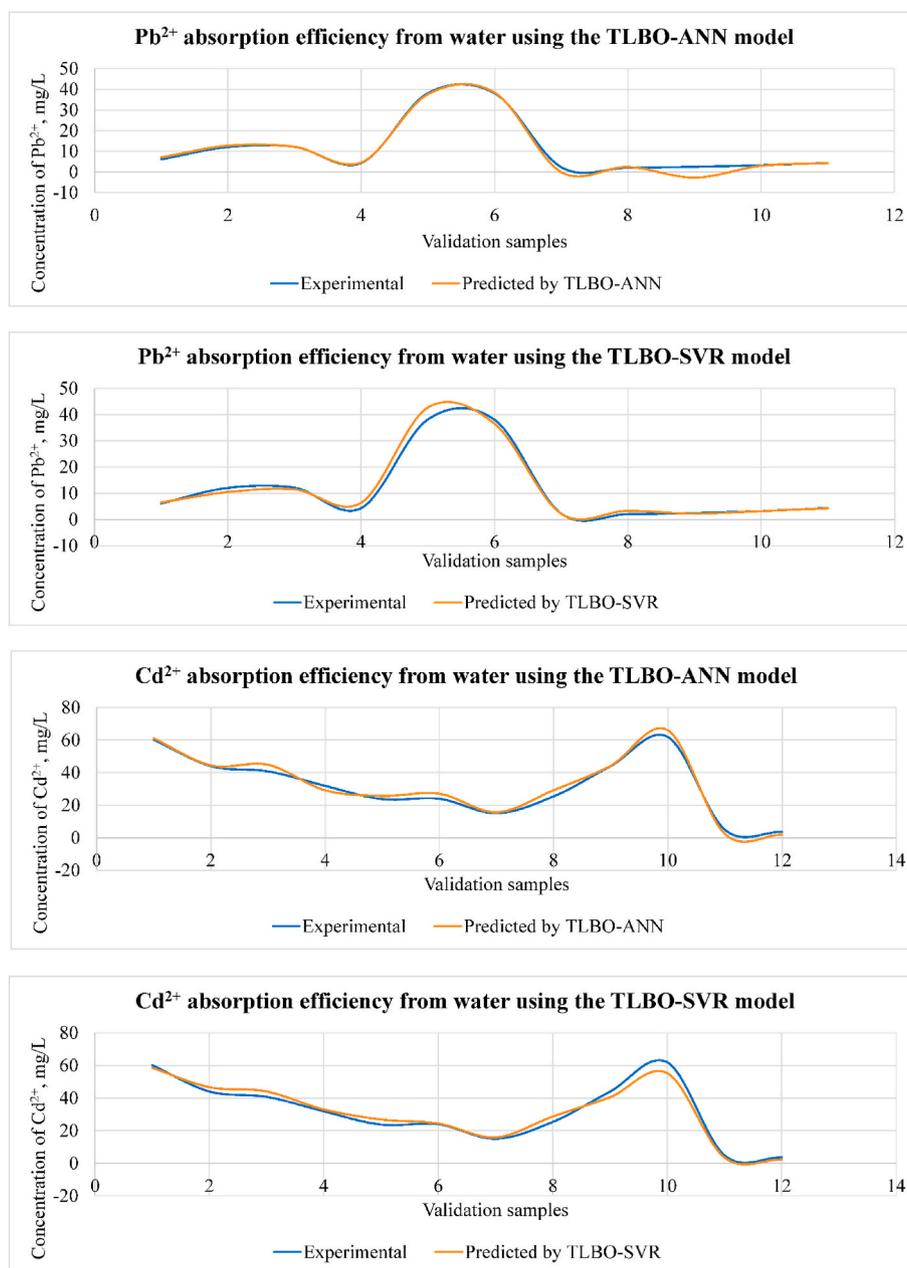


Fig. 7. Results and accuracies of the TLBO-ANN and TLBO-SVR models. in the validation dataset.

Table 6

Performance of the TLBO-ANN and TLBO-SVR models in the validation dataset.

Heavy metals absorption	Models	Validation dataset		
		RMSE	R ²	VAF
Pb ²⁺ absorption	TLBO-ANN	1.785	0.985	98.300
	TLBO-SVR	1.709	0.986	98.366
Cd ²⁺ absorption	TLBO-ANN	2.585	0.988	98.323
	TLBO-SVR	2.929	0.975	97.464

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

This research was financially supported by the Vietnam National Foundation for Science and Technology Development (NAFOSTED), under grant number 105.99-2017.317.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.chemosphere.2021.131012>.

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