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Adsorption-based sustainable recovery of heavy metals from water resources by secondary raw materials

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Summary

Adsorption-based processes using waste-derived materials have emerged as a promising and sustainable approach for the removal of heavy metals from contaminated water resources, particularly in response to increasing environmental pressure and the need for circular economy-oriented treatment strategies. Among various secondary raw materials, construction and demolition waste, and specifically Low-Density Concrete (LDC), offers high potential due to its cementitious composition, porosity, and chemical stability, enabling effective immobilization of toxic metal ions such as Pb^{2+} , Co^{2+} , and Mn^{2+} .

Although numerous studies have demonstrated the feasibility of waste-based adsorbents for heavy metal removal, the exact adsorption mechanisms, kinetic and equilibrium behavior, and environmental performance of LDC remain insufficiently understood. Furthermore, conventional adsorption systems are typically operated empirically, with limited automation and weak integration of predictive modeling, optimization, and process supervision, which restricts their scalability and industrial applicability. Consequently, detailed experimental investigation combined with intelligent decision-support strategies is required to bridge laboratory-scale adsorption studies with real-world water treatment applications.

To address these challenges, this thesis systematically investigated the adsorption of Pb^{2+} , Co^{2+} , and Mn^{2+} onto LDC through mechanistic analysis, isothermal and kinetic modeling, and sustainability assessment. Experimental results demonstrate that adsorption occurs predominantly via physical interactions on heterogeneous surfaces, strongly influenced by pH, metal type, and adsorbent dosage. To extend beyond static performance evaluation, an AI-based decision-making framework was developed by integrating Artificial Neural Networks, Genetic Algorithm optimization, and Monte Carlo uncertainty analysis. This framework enabled accurate prediction of adsorption performance, identification of metal-specific optimal operating conditions, and quantification of operational robustness under uncertainty.

By combining waste valorization with intelligent automation, the developed decision-support system transforms adsorption from a static treatment technique into an adaptive, data-driven process capable of supporting informed operational decisions. Accordingly, this thesis advances the understanding of heavy metal adsorption mechanisms on waste-derived LDC and contributes to the development of sustainable, intelligent, and scalable water treatment strategies, providing a practical foundation for future Industry 4.0-oriented adsorption systems in complex and variable aqueous environments.

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1. Introduction

Heavy metal concentrations in wastewater have increased as a result of human and industrial processes like tanning, mining, petrochemicals, textiles, and plating. Because these metals are frequently carcinogenic and non-biodegradable, tainted water can enter the environment and pose a serious risk to human health and ecosystems [1]. The growing regulatory attention to heavy metal contamination in water, reflected in the progressive tightening of European Union drinking water standards under Directive (EU) 2020/2184, further underscores the critical importance of developing effective and sustainable removal technologies capable of meeting increasingly stringent water quality requirements [2].

Adsorption stands out as one of the most effective and extensively studied heavy metal removal strategies. It is the process by which metal ions, which are usually obtained from inexpensive or waste materials like natural minerals, industrial byproducts, or agricultural residues, adhere to solid surfaces. Its primary benefits include cost-effectiveness, environmental compatibility, high removal efficiency, and ease of operation. Adsorption also converts plentiful waste materials into useful resources, which is consistent with the ideas of the circular economy. However, adsorbent saturation causes its efficiency to decline with repeated use, and regeneration may necessitate chemical treatments that produce secondary waste. Additionally, adsorbent selectivity for particular metals is restricted, and it is still difficult to maintain reliable performance in large-scale operations. Furthermore, adsorption systems' limited automation and control programming point to a process optimization weakness. Adsorption is nevertheless a viable and sustainable method of treating wastewater in spite of these disadvantages [3].

For heavy metal decontamination, a variety of adsorbents are used, such as membrane-based [4], mineral [5], magnetic [6], bio- [7], and polymer-based [8] sorbents. Because of their abundance, affordability, and environmental advantages, waste materials-based sorbents have drawn particular attention among them. The Sustainable Development Goals (SDGs) [9], especially those pertaining to waste reduction, clean water, and responsible consumption, are substantially supported by the use of waste-derived adsorbents. The Integrated Solid Waste Management (ISWM) concept [10], which turns environmental burdens into useful adsorptive materials by emphasizing resource recovery and circular usage of waste streams, is the source of this alignment.

Waste-based adsorbents (WBAs) provide a workable way to reduce decontamination expenses and encourage coordination between water treatment and solid waste management programs. WBAs have drawn interest recently as an economical and environmentally responsible way to rid water sources of dangerous contaminants, especially heavy metals. Because of their high adsorption efficiency, low toxicity, and affordability, a variety of natural materials have been used as green adsorbents, including leaves, walnut shells, coir pith, clay, and agricultural residues. Furthermore, the range of sustainable wastewater treatment options has been further expanded by the successful adsorption of heavy metals from contaminated water by waste-derived inorganic materials like bentonite, dolomite, and zeolite [10].

Construction and Demolition Wastes (CDWs) are valuable secondary resources for environmental remediation, especially in heavy metal decontamination. Their cementitious composition and high surface reactivity enable effective adsorption of toxic ions such as Pb^{2+} , Cd^{2+} , and Zn^{2+} from contaminated water. Utilizing CDWs not only addresses pollution control but also promotes circular economy practices by converting waste into useful adsorbent materials [11]. Among all different types of CDWs, Low Density Concrete (LDC), as a prominent component of CDWs, exhibits a high porosity level, which significantly enhances its adsorption capacity. This porous network increases the available surface area for ion exchange and diffusion, allowing heavy metals to be effectively trapped within the matrix. The lightweight nature and chemical stability of LDC make it a sustainable, low-cost material ideal for large-scale heavy metal removal and wastewater treatment applications [10].

In the following sections, the state of the art, research objectives, results and discussions, conclusion, outline of future works, and academic achievements are presented.

2. State of the art

2.1. CDW application for adsorption of heavy metals

Materials produced from C&D waste have been extensively investigated for the removal of heavy metals. Because of its alkaline and calcium-rich nature, waste cement powder has been used to adsorb As^{5+} ions from aqueous environments [12]. Likewise, in the other study, As^{5+} is efficiently captured by the concrete/maghemite nanocomposite (*CMnano*) via surface complexation and magnetically aided adsorption [13]. Because it contains active iron sites that promote adsorption, vermiculite concrete treated with ferric oxyhydroxide effectively adsorbs Cu^{2+} , Cd^{2+} , and Pb^{2+} [14]. Crushed concrete fractions (CCF) have also been used to remove Pb^{2+} , Cu^{2+} , and Zn^{2+} through precipitation and adsorption, offering a low-cost and environmentally friendly method of wastewater treatment [15].

Wijeyawardana et al. [16] used biochar modified with Ordinary Portland Cement (OPC) as an adsorbent for Cu^{2+} , Pb^{2+} , and Zn^{2+} removal. The OPC-modified biochar showed 3.3×, 2.4×, and 6× higher adsorption than unmodified biochar. Competing ions (NO_3^- , PO_4^{3-} , NH_4^+) and humic acid effects were evaluated for adsorption performance. The processes by which the cement–biochar composite removes Cu^{2+} , Pb^{2+} , and Zn^{2+} are depicted in Fig. 1. The porous microstructure of the composite is seen in the center SEM micrograph, which is encircled by

schematic illustrations of the adsorption and precipitation processes. Sections C and D show ion exchange between Ca^{2+} in calcium silicate hydrate (C–S–H) and metal ions, as well as adsorption on silanol/siloxane (Si-OH , Si-O-Si) sites. Sections A–B show complexation of metal ions with C=O and -COOH groups. While G emphasizes surface adsorption on aromatic or aliphatic sites, Sections E–F demonstrate hydroxide and carbonate precipitation. These interactions collectively show that chemical bonding, ion exchange, and precipitation processes work in concert to remove heavy metals [16].

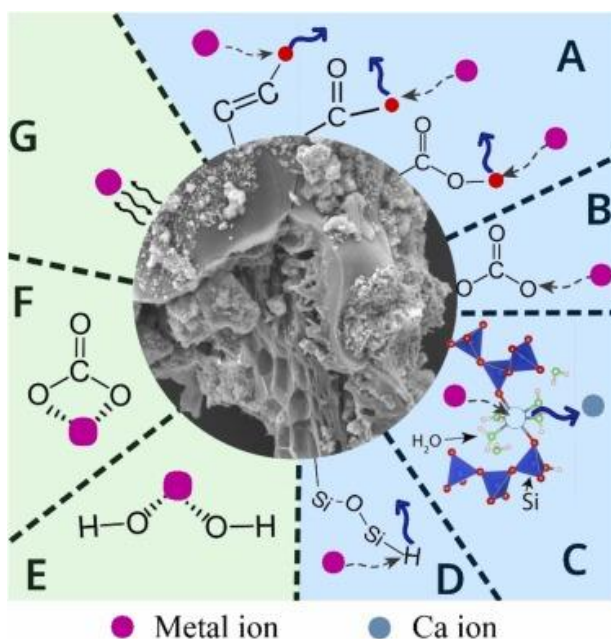


Fig. 1. Schematic illustration of Cu^{2+} , Pb^{2+} , and Zn^{2+} removal by cement–biochar composite through functional group complexation, Ca^{2+} ion exchange, and precipitation of metal hydroxides and carbonates, reference: [16].

Concrete sludge is a slurried industrial byproduct composed of aggregates, hydrated cement, and water. Yoo et al. [17] investigated the adsorption of Pb^{2+} , Cu^{2+} , and Zn^{2+} ions using dried concrete sludge with a fine particle size ($<25\ \mu\text{m}$), which was mainly composed of muscovite ($\text{KAl}_2(\text{Si}_3\text{AlO}_{10})(\text{OH})_2$), quartz (SiO_2), and albite ($\text{NaAlSi}_3\text{O}_8$). Their study demonstrated the high effectiveness of this material in removing these heavy metals over an extended contact period. Similarly, using demolished concrete (1–2 mm particle size) as an adsorbent, Ali and Abd Ali [18] carried out a similar study to extract Pb^{2+} , Cu^{2+} , and Ni^{2+} ions from synthetic wastewater. The material's adsorption capability was evaluated by both batch and column studies. The investigation showed that within a moderate contact period, demolished concrete demonstrates a substantial adsorption ability for these heavy metal ions.

2.2. Mechanism of heavy metal adsorption onto CDWs

Each adsorption process involves two key aspects: the adsorption strength, determined through isotherm analysis, and the reaction rate, derived from kinetic analysis [19]. Accordingly, the detailed mechanism of heavy metal adsorption onto CDW can be comprehensively examined.

2.2.1. Isotherm models

The most recent developments in the isothermal investigation of heavy metal adsorption onto different CDWs are compiled in Table 1, which highlights the fundamental adsorption mechanisms. Monolayer chemisorption on homogeneous surfaces and multilayer physisorption on heterogeneous ones are the two predominant adsorption processes represented by the Langmuir and Freundlich/Dubinin–Radushkevich models. In line with these mechanisms, biochar-admixed mortar fines exhibit significant adsorption capacity ($\text{Pb}^{2+} = 476.19 \text{ mg g}^{-1}$), attributed to the oxygenated surface groups introduced by biochar [20].

The Langmuir behavior of clay brick, aerated concrete and ceramic sorbents indicates homogeneous active sites and predominant surface complexation for heavy metal binding [21], [22]. On the other hand, heterogeneous surface adsorption caused by van der Waals forces and electrostatic interactions are revealed in Freundlich-type systems including washed cement paste, lightweight concrete, and brick, with mean adsorption energies ($E = 4\text{--}5 \text{ kJ mol}^{-1}$) corroborating physisorption [23]. Because porous brick and cement phases coexist, construction waste mixes have synergistic effects that improve multilayer adsorption and metal ion diffusion [24]. Overall, Table 1 shows how modifying CDWs' surface chemistry, pore shape, and mineral composition controls their adsorption mechanism and effectiveness, making them viable, affordable options for heavy-metal cleanup.

Table 1. The summary of isothermal analysis for adsorption of heavy metals onto CDWs.

Adsorbent	Adsorbate	Isothermal model	Isothermal parameters	Reference
Biochar-admixture cement mortar fines (40 % of sand replaced by biochar)	Pb^{2+} , Cu^{2+} , and Zn^{2+}	Langmuir	Pb^{2+} ($q_{\text{max}}=476.19 \text{ mg.g}^{-1}$, $K_L=0.43 \text{ L.mg}^{-1}$), Cu^{2+} ($q_{\text{max}}=80.64 \text{ mg.g}^{-1}$, $K_L=0.75 \text{ L.mg}^{-1}$), and Zn^{2+} ($q_{\text{max}}=123.45 \text{ mg.g}^{-1}$, $K_L=1.4 \text{ L.mg}^{-1}$)	[20]
Ceramic sorbent (CS)	Pb^{2+} , Cu^{2+} , and Zn^{2+}	Langmuir	Pb^{2+} ($q_{\text{max}}=186 \text{ mg.g}^{-1}$), Cu^{2+} ($q_{\text{max}}=91 \text{ mg.g}^{-1}$), and Zn^{2+} ($q_{\text{max}}=43 \text{ mg.g}^{-1}$)	[21]
Aerated Concrete (AC)	Pb^{2+}	Langmuir	184.9 mg.g^{-1}	[22]
Clay Brick (CB)	Pb^{2+}	Langmuir	42.9 mg.g^{-1}	[22]
Cement paste (washed)	Cd^{2+}	Freundlich and Dubinin-Radushkevich models	$E=5.08 \text{ kJ.mol}^{-1}$; $K_F=689 \text{ L.kg}^{-1}$; $1/n=0.780$	[23]

Lightweight concrete (washed)	Cd ²⁺	Freundlich and Dubinin-Radushkevich models	E=4.18 kJ·mol ⁻¹ ; K _f =130 L·kg ⁻¹ ; 1/n=0.677	[23]
Brick	Cd ²⁺	Freundlich and Dubinin-Radushkevich models	E=4.32 kJ·mol ⁻¹ ; K _f =167 L·kg ⁻¹ ; 1/n=0.703	[23]
Construction waste (mainly brick with a small amount of concrete, 2.36–4.75 mm particle size)	Pb ²⁺ , Cu ²⁺ , and Cd ²⁺	Freundlich	Cu ²⁺ (K _f = 0.181 ± 0.053 L·kg ⁻¹ ; 1/n= 0.785 ± 0.24) Cd ²⁺ (K _f = 0.106 ± 0.031 L·kg ⁻¹ ; 1/n= 1.065 ± 0.55) Pb ²⁺ (K _f = 37.58 ± 1.58 L·kg ⁻¹ ; 1/n= 1.402 ± 0.52)	[24]

2.2.2. Kinetics models

State-of-the-art evidence shows that CDWs exhibit highly variable adsorption capacities and kinetic behaviors depending on mineralogy, particle size, and the dominance of surface precipitation versus true sorption (Table 2). For arsenate removal, waste cement powder reaches an adsorption capacity of 28.3 mg g⁻¹, but requires a very long equilibrium/contact time (1440 min) under strongly alkaline conditions (pH 10–11), implying slow diffusion and/or gradual formation of stable binding/precipitated phases rather than fast site-limited uptake [12]. In contrast, a *CMnano* shows a lower As⁵⁺ capacity (11.12 mg g⁻¹) and still slow uptake (2400 min) at pH 5, yet its kinetics follow Pseudo-Second-Order (PSO) behavior with a reported rate coefficient of 0.008 g mg⁻¹ min⁻¹, indicating that chemisorption-like surface reactions (complexation on iron-active sites) likely control the rate despite the extended time to reach equilibrium [13]. For cationic metals, cementitious CDWs can achieve much higher capacities: vermiculite concrete modified with ferric oxyhydroxide removes Pb²⁺ with a capacity of 197.6 mg g⁻¹ and a dramatically shorter optimal contact time (120 min) at pH 5, also fitting PSO kinetics with a markedly higher rate coefficient (0.299 g mg⁻¹ min⁻¹), consistent with abundant reactive hydroxyl/iron sites enabling fast surface complexation and possible co-precipitation [14]. By comparison, CCF shows a moderate Pb²⁺ capacity (37.9 mg g⁻¹) but requires very long contact times (2880 min) at high pH (11.9), suggesting slower mass transfer and a stronger reliance on alkaline precipitation mechanisms and intraparticle diffusion [15]. In general, CDWs span fast-high-capacity modified systems to slow-moderate-capacity unmodified fractions, and the coupling between adsorption capacity and kinetic order/rate constant is strongly material-dependent rather than universal. Given LDC's high porosity and cementitious chemistry highlighted in this thesis, a dedicated kinetic capacity mapping (e.g., PSO/PFO/intraparticle diffusion fitting across pH and dosage) would be a logical next step to rigorously analyze how LDC compares to the CDWs above in both adsorption capacity and rate-limiting mechanisms.

Table 2. Summary of studies on the application of construction and demolition waste (CDW) as an adsorbent for heavy metal decontamination, with a focus on adsorption kinetics and capacity.

Adsorbent	Contamination	Adsorption capacity (mg.g ⁻¹)	Optimal contact time (min)	Operational pH	Kinetics order	Kinetics Coefficient (g.mg ⁻¹ .min ⁻¹)	Ref.
Waste cement powder	As ⁵⁺	28.3	1440	10-11	_____	_____	[12]
<i>CMnano</i>	As ⁵⁺	11.12	2400	5	PSO	0.008	[13]
Vermiculite concrete modified with Ferric oxyhydroxide	Pb ²⁺	197.6	120	5	PSO	0.299	[14]
CCF	Pb ²⁺	37.9	2880	11.9	_____	_____	[15]

Research Gap (RG) 1 is defined as the lack of a clear chemical understanding of the mechanisms governing heavy metal adsorption onto LDC. This research gap will be met in this thesis by elucidating the dominant adsorption pathways through isothermal modelling, pH-controlled experiments, and surface characterization. The study systematically evaluates the roles of physisorption on heterogeneous surfaces, Ca²⁺/Al³⁺ ion exchange, and complexation with surface hydroxyl groups, primarily associated with C–S–H and occurring on the surfaces of Si–O and Al–O phases.

RG2 is defined as the absence of comprehensive information on the isothermal and kinetic behavior of heavy metal adsorption on LDC. This gap will be addressed by applying multiple isotherm models to identify adsorption regime and surface heterogeneity (Freundlich and Langmuir models), while Dubinin–Radushkevich analysis clarifies the nature of interactions. Kinetic modelling is used to determine adsorption order and rate-limiting steps, linking adsorption dynamics to active site availability, porosity, and the Al₂O₃-enriched cementitious structure of LDC.

2.3. Environmental Impact Assessment of adsorbents

Environmental impact assessment of adsorbents is increasingly conducted using cradle-to-gate Life Cycle Assessment (LCA) frameworks integrating the Tool for the Reduction and

Assessment of Chemicals and Other Environmental Impacts and the ReCiPe endpoint methodology. Recent studies emphasize performance-normalized functional units based on adsorption capacity and specific surface area to decouple material efficiency from mass-based impacts. Energy consumption, chemical activation, and scalability dominate midpoint and endpoint indicators, demonstrating the need to couple adsorption kinetics with sustainability assessment [25]. In the following, Wang et al. [26] applied life cycle assessment to algae-derived carbon adsorbents, linking adsorption efficiency with preparation impacts. Their results show that adsorbent yield, activation chemistry, and pore development strongly influence environmental performance, with Enteromorpha-based adsorbents achieving high tetracycline removal at lower life-cycle burdens. Plus, Kazemi et al. [27] applied a prospective cradle-to-use Life Cycle Assessment to nanoadsorbents for mercury removal, using 1 kg Hg removal as the functional unit. Their results showed that adsorbent functionalization significantly increases adsorption capacity but also amplifies cumulative energy demand, human toxicity, and ecotoxicity, identifying electricity use and coupling reagents as dominant environmental hotspots. Whereas Gonzalez et al. [28] performed a cradle-to-use LCA of silicate–titanate nanotube–chitosan nanoadsorbents for cadmium removal. Their analysis demonstrated that despite high adsorption capacity and recyclability, nanoadsorbent synthesis is dominated by electricity demand and alkaline activation, making energy efficiency and regeneration critical for sustainable scale-up when compared with commercial activated carbon.

RG3 originates from the inadequate incorporation of CDW-derived adsorbents, such as LDC, into environmental impact assessment frameworks. Existing studies largely prioritize virgin or engineered adsorbents, applying mass-based life cycle metrics that overlook waste valorization, avoided disposal, regeneration benefits, and adsorption-normalized functional units. Moreover, alignment with the U.S. Environmental Protection Agency Sustainable Materials Management (SMM) framework, emphasizing resource efficiency and material circularity, remains largely absent in adsorption-related sustainability analyses.

2.4. Automation and AI applications in adsorption of heavy metals

Recent advances in Artificial Intelligence (AI) have enabled the use of Machine Learning Algorithms (MLAs) to accurately predict, optimize, and automate heavy metal adsorption processes by capturing the complex nonlinear interactions between adsorbent properties and

operational parameters. Table 3 highlights the growing integration of machine learning algorithms as data-driven tools for modeling and automating heavy metal adsorption processes across diverse adsorbent classes. Advanced ensemble learning methods, particularly Gradient Boosting-based algorithms, dominate recent studies due to their strong nonlinear learning capability and robustness against multicollinearity [29] in adsorption datasets. Metal-organic frameworks [29] and hydrochars [30] have been successfully modeled using Gradient Boosting Decision Trees, achieving high predictive accuracy (R^2 up to 0.95) for multi-metal systems, indicating their suitability for complex competitive adsorption scenarios. Random Forest models applied to biochar systems demonstrate excellent generalization performance ($R^2 = 0.973$) [31], benefiting from inherent feature bagging and resistance to overfitting. Extreme Gradient Boosting (XGBoost) exhibits the highest reported accuracy across bentonite [32] and clay mineral adsorbents [33] (R^2 up to 0.95), reflecting its effectiveness in capturing subtle interactions between process variables such as pH, dosage, and initial concentration. Generally, these studies confirm that tree-based ensemble learning models outperform conventional statistical approaches in adsorption prediction, enabling real-time optimization, process automation, and intelligent decision support. However, most existing applications remain limited to prediction tasks, with minimal integration into closed-loop control systems or sustainability-oriented process automation, highlighting a critical gap addressed in this study.

Table 3. Overview of selected studies investigating the use of machine learning algorithms in adsorption processes.

Adsorbent	Adsorbate	Machine learning model	Statistical indicators	Reference
89 metal-organic frameworks composites	As ^{3+/5+} , Cd ²⁺ , Cr ⁶⁺ , Cu ²⁺ , Pb ²⁺ , Hg ²⁺	Gradient Boosting Decision Tree (GBDT)	$R^2=0.921-0.962$ for 6 heavy metals	[29]
Biochar	Cu ²⁺ , Zn ²⁺ , Pb ²⁺ , Cd ²⁺ , Ni ²⁺ , As ³⁺	Random forest	$R^2 = 0.973$	[31]

Bentonite	Pb ²⁺ , Cu ²⁺ , Cd ²⁺ , Zn ²⁺ , Ni ²⁺ , Co ²⁺ , and chromium ions	eXtreme Gradient Boosting Regression (XGB)	Train $R^2 = 0.995$ Test $R^2 = 0.9359$	[32]
Hydrochar	Pb ²⁺ , Cd ²⁺ , Zn ²⁺ , Cu ²⁺ , Sb ³⁺ , Sb ⁵⁺ , CrO ₄ ²⁻ , UO ₂ ²⁺	Gradient boosting decision tree	$R^2 = 0.93-0.98$	[30]
Clay minerals	Cd ²⁺ , Cu ²⁺ , Pb ²⁺ , Zn ²⁺ , Ni ²⁺ and Co ²⁺	Extreme Gradient Boosting	$R^2 = 0.977$	[33]

RG4 relates to the absence of an integrated automation strategy for heavy metal adsorption onto LDC that extends beyond prediction and regression-based model extension. Existing machine learning applications predominantly focus on forecasting adsorption performance, while lacking mechanisms for closed-loop control, alarm management, and degradation-aware supervision. In particular, current models do not address real-time detection or control of adsorbent performance loss during decontamination processes. This limitation restricts their applicability in dynamic and automated operational environments.

According to the literature review and evaluation of previous studies in the field of CDW, particularly LDC used as an adsorbent for heavy metals, four RGs have been identified as follows:

RG1: *The exact mechanism of heavy metal adsorption onto LDC.*

RG2: *The isothermal and kinetic behavior of heavy metal adsorption on the surface of LDC.*

RG3: *The environmental impacts of LDC application compared to other adsorbents.*

RG4: *The appropriate method for automating the heavy metal adsorption process onto LDC using AI models.*

2.5. Limitations

For the decontamination of heavy metals from water and wastewater treatment systems, several treatment methods have been developed, among which adsorption is recognized as a particularly effective and versatile technique [3]. Despite its advantages, adsorption still exhibits notable limitations, as illustrated in Fig. 2, including a relatively low level of automation, insufficient environmental performance assessment, and challenges in quantitative evaluation and process control [3]. Therefore, based on the classical principles of adsorption, such as elucidating

the underlying mechanisms governing heavy metal uptake onto the surface of CDWs including LDC, the present thesis focuses on a more integrated and systematic approach. Specifically, the SMM model was applied to compare and quantify the green performance of different adsorbents. In parallel, AI models encompassing prediction, classification, and optimization were implemented within a smart expert system framework to address existing gaps, enhance decision-making capability, and comprehensively evaluate the technical, environmental, and operational aspects of the adsorption process.

Methods of wastewater treatment

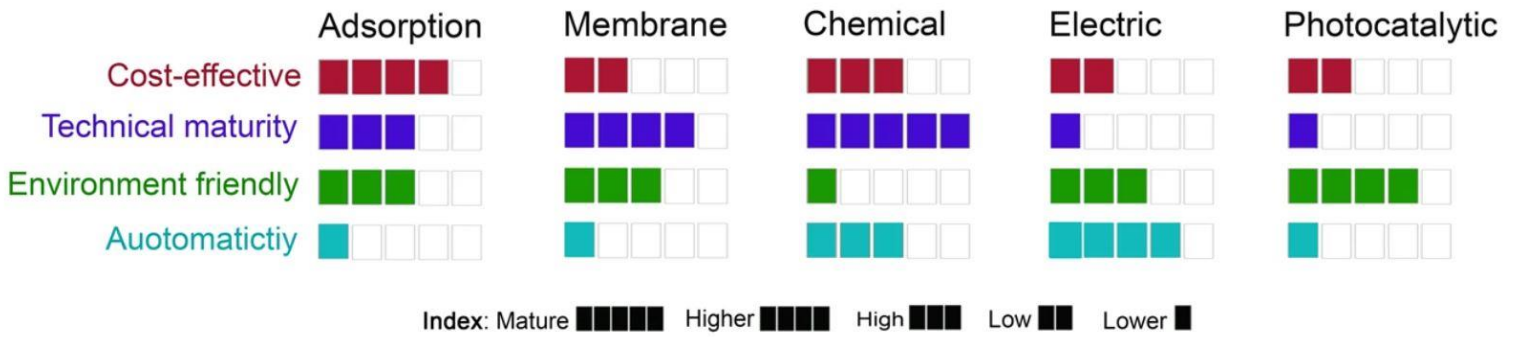


Fig. 2. The adsorption-, membrane-, chemical-, electric-, and photocatalytic-based methods are compared with respect to cost-effectiveness, technical maturity, environmental friendliness, and level of automation, based on the preceding discussion and evidence reported in the literature [3].

Therefore, the objectives of this thesis are defined to include mechanistic analysis, isothermal and kinetic assessments, environmental impact evaluation of the waste-based LDC adsorbent, and the automation of the heavy metal adsorption process onto LDC.

Fig. 3 (a-f) presents a comprehensive multi-metric evaluation of adsorbents, clearly highlighting the performance significance of the low-cost derived composite (LDC). In Fig. 3a, the Cleveland dot plot shows that LDC (Pb^{2+}) achieves the highest adsorption capacity (43.10 mg g^{-1}) [10], outperforming conventional materials such as Meranti sawdust (35.97 mg g^{-1})[34] and Autoclaved Aerated Concrete (AAC) (16.50 mg g^{-1})[35], while maintaining competitive performance for Co^{2+} (15.17 mg g^{-1})[10] and Mn^{2+} (23.52 mg g^{-1}) [10]. This confirms the superior adsorption behavior of LDC reported in the present study. Fig. 3(b) correlates capacity with surface area, revealing that despite silica ceramic having an extremely high surface area ($340.38 \text{ m}^2 \text{ g}^{-1}$) [36], [37], its adsorption capacity remains low (2.7 mg g^{-1})[36], [37], indicating that surface area alone does not govern performance. In contrast, LDC achieves high capacity at moderate surface

area ($30.2 \text{ m}^2 \text{ g}^{-1}$)[10], suggesting efficient active site utilization. This is further emphasized in Fig. 3(c), where LDC (Pb^{2+}) is positioned in the “high capacity-high performance” quadrant, demonstrating optimal balance between adsorption efficiency and material structure. The normalized comparisons in Fig. 3(d-e) reinforce this trend, where LDC exhibits consistently high scores in capacity and efficiency metrics, unlike other materials that show trade-offs. The spider chart in Fig. 3(f) confirms LDC’s balanced multi-metric profile, particularly for Pb^{2+} removal. Overall, these results align with previous findings [34], [35] while demonstrating that the LDC developed in this study offers a more efficient and scalable adsorption solution, making it a key material within this thesis.

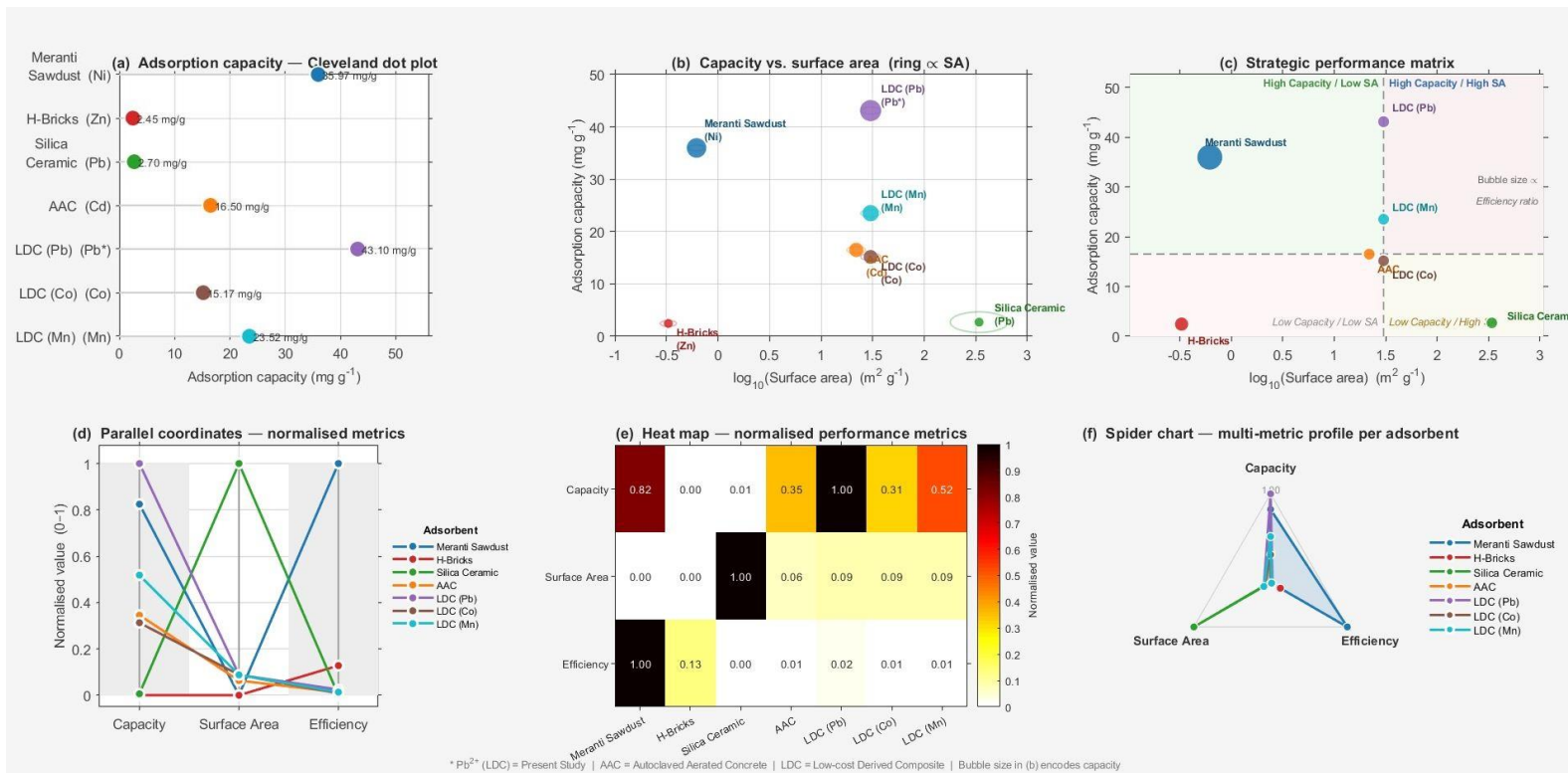


Fig. 3. The comparison between different CDW adsorbents for heavy metal decontamination as per: (a) Adsorption capacity comparison in Cleveland dot plot, (b) Capacity vs. surface area, (c) Strategic matrix placing, (d) Normalized metrics comparison, (e) Heat map of performance, and (f) Spider chart.

Based on the four identified research gaps, the following research objectives were systematically formulated to address both the scientific and technological deficiencies in LDC-based heavy metal adsorption, ensuring that each objective directly responds to a specific gap in existing knowledge.

3. Thesis objectives

The present work aims to overcome existing scientific and technological gaps related to the use of LDC waste as a sustainable adsorbent for heavy metal removal from water. The detailed research contents and objectives are structured based on the identified RGs and are listed below.

RG1, from which **Research Objective 1** of this thesis was derived, highlights the insufficient understanding of the exact physicochemical mechanisms governing heavy metal adsorption onto LDC. Accordingly, **Research Objective 1** focuses on clarifying these mechanisms. The specific research objectives are:

- A1.** to identify the dominant adsorption mechanisms involved in the removal of Pb^{2+} , Co^{2+} , and Mn^{2+} onto LDC surfaces,
- A2.** to investigate the role of surface functional groups, cementitious phases, and mineral composition of LDC in metal uptake,
- A3.** to evaluate the contribution of physisorption, ion exchange, and surface complexation processes in heavy metal immobilization,
- A4.** to elucidate the influence of solution pH on adsorption pathways and metal-surface interactions.

These objectives collectively aim to move beyond the qualitative descriptions found in existing CDW literature toward a mechanistically grounded and experimentally validated understanding of how LDC surface chemistry governs metal uptake.

RG2, from which **Research Objective 2** was derived, highlights the lack of comprehensive knowledge regarding the isothermal and kinetic behavior of heavy metal adsorption on LDC. Accordingly, **Research Objective 2** focuses on addressing this gap. Thus, the specific research objectives are:

- B1.** to determine the adsorption equilibrium behavior of Pb^{2+} , Co^{2+} , and Mn^{2+} onto LDC using multiple isotherm models,
- B2.** to assess surface heterogeneity and adsorption intensity through comparative isothermal analysis,
- B3.** to evaluate the adsorption kinetics and identify the governing rate-limiting steps of the process,
- B4.** to correlate kinetic and isothermal parameters with the structural and porosity characteristics of LDC.

Together, these objectives provide the quantitative foundation needed to link LDC's structural and mineralogical properties to its practical adsorption performance, enabling reliable prediction under variable water quality conditions.

RG3, from which **Research Objective 3** was derived, highlights the need to evaluate the environmental performance of LDC-based adsorption systems in comparison with conventional and engineered adsorbents. Accordingly, **Research Objective 3** focuses on this evaluation. The specific research objectives are:

- C1.** to assess the environmental impacts of LDC application as an adsorbent using SMM and life-cycle-based approaches,
- C2.** to compare the environmental performance of LDC with that of conventional adsorbents using adsorption-normalized functional units,
- C3.** to identify key environmental hotspots associated with LDC preparation, application, regeneration,
- C4.** to quantify the sustainability benefits of waste valorization and avoided disposal within the adsorption process.

This objective extends the scope of the thesis beyond technical performance, recognizing that a waste-derived adsorbent can only be justified for large-scale adoption if its environmental burden is demonstrably lower than or comparable to that of conventional alternatives across the full material life cycle.

RG4, from which **Research Objective 4** was derived, highlights the limited automation and intelligence of conventional adsorption processes, particularly the lack of integration of artificial intelligence-based control strategies. Accordingly, **Research Objective 4** focuses on overcoming these limitations. Thus, the specific research objectives are:

- D1.** to develop machine learning models capable of accurately predicting heavy metal adsorption performance under nonlinear operating conditions,
- D2.** to optimize adsorption operating parameters using AI-based optimization techniques to maximize removal efficiency,
- D3.** to assess process robustness and uncertainty using stochastic simulation methods,
- D4.** to design an AI-based expert system enabling automated supervision, performance degradation detection, and intelligent decision-making for heavy metal adsorption onto LDC.

These objectives collectively transform the adsorption system from a passive, experiment-driven process into an active, data-informed operational framework, where AI models serve not merely as predictive tools but as the computational core of an intelligent supervision and control architecture aligned with Industry 4.0 principles.

4. Results and discussions

As the first author, I published a research paper in a **Q1** journal (impact factor: 7.7) entitled “*Heavy Metals Adsorption Using CDW Adsorbents: A Sustainable Path for Water Purification*”, in which **Research Objectives 1-3** were achieved. In the subsequent stage, another paper was accepted in a **Q2** journal (impact factor: 4.1) entitled “*Smart Control of Heavy Metal Adsorption onto LDC Wastes for Industry 4.0 Applications*”, which addresses **Research Objective 4**. Finally, **Research Objective 4** was further fulfilled through a paper published in the **Q3** journal *Chemistry–Didactics–Ecology–Metrology* (impact factor: 1.2), entitled “*AI-Based Expert System Design for Optimisation of Heavy Metal Adsorption on Waste-Derived Adsorbents*”.

4.1. Heavy metals adsorption using CDW adsorbents

Efficient elimination of heavy metals from water is vital for safeguarding both human health and the environment, and adsorbents offer a sustainable and economical treatment approach. This research presents the first investigation into the use of LDC as an adsorbent for the removal of specific heavy metals (Pb^{2+} , Co^{2+} , and Mn^{2+}) from water. The material was characterized through X-ray fluorescence (XRF), Fourier Transform Infrared Spectroscopy (FTIR), Brunauer–Emmett–Teller (BET) analysis, X-ray diffraction (XRD), and Field Emission Scanning Electron Microscopy (FE-SEM). The One-Factor-at-a-Time (OFAT) approach was applied to optimize the key operational parameters influencing metal adsorption. Eight distinct isotherm models were employed to estimate adsorption parameters and clarify the underlying sorption mechanisms, followed by kinetic and selectivity analyses for decontamination. The results revealed adsorption capacities of 43.1, 23.5, and 15.2 mg g^{-1} for Pb^{2+} , Mn^{2+} , and Co^{2+} , respectively. The corresponding second-order kinetic rate constants were 1.99, 0.076, and 0.49 $\text{g mg}^{-1} \text{ min}^{-1}$. As an innovative aspect, the regeneration of the adsorbent was modeled using a Thales-based theorem. Synthetic wastewater was used for experimentation, while regeneration conditions were designed to replicate industrial effluent scenarios, enhancing practical relevance. Additionally, the use of CDW-based LDC was assessed under the Sustainable Material Management framework using EPA models.

Isothermal analysis indicated that adsorption of Pb^{2+} , Co^{2+} , and Mn^{2+} onto LDC occurred via physical interactions on a heterogeneous surface through multilayer adsorption. The Thales-based model further showed that after nine adsorption–regeneration cycles with acid treatment, the LDC adsorbent consistently reduced lead concentrations to below the World Health Organization (WHO) permissible limit. The main aspects of this section are summarized below. Existing studies adequately address **Research Objectives 1-3**.



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Heavy metals adsorption using CDW adsorbents: A sustainable path for water purification

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ABSTRACT

Effective removal of heavy metals from water is crucial for human health and environmental protection, and adsorbents provide a sustainable, cost-effective treatment solution. This study is the first to investigate Low-Density Concrete (LDC) for removing selected heavy metals (Pb^{2+} , Co^{2+} , and Mn^{2+}) from water. The adsorbent was characterised using X-ray fluorescence (XRF), Fourier Transform Infrared spectrophotometry (FTIR), Brunauer-Emmett-Teller (BET) analysis, X-ray diffraction (XRD) analysis, and Field Emission Scanning Electron Microscopy (FE-SEM). The One-Factor-at-A-Time (OFAT) method optimised the essential parameters affecting the metal removal. Eight different isotherm models were used to determine the adsorption parameters and elucidate the associated sorption mechanism. Then, its kinetic behavior and selectivity for decontamination were analyzed. The outcomes demonstrated that the adsorption capacities for Pb^{2+} , Mn^{2+} , and Co^{2+} were 43.1, 23.5, and 15.2 mg g^{-1} , respectively. Moreover, the second-order kinetic rate constants for Pb^{2+} , Mn^{2+} , and Co^{2+} were determined to be 1.99, 0.076, and 0.49 $\text{g mg}^{-1} \text{min}^{-1}$, respectively. As a novel contribution, the regeneration process of heavy metal adsorption is modelled by Thales-based theorem. In this study, synthetic wastewater was used, while regeneration conditions simulated industrial effluents to enhance real-world relevance. Finally, the applied construction and demolition waste (CDW) adsorbent is evaluated by Sustainable Material Management, EPA models. The isothermal modelling illustrated that the adsorption of Pb^{2+} , Co^{2+} , and Mn^{2+} onto LDC wastes is done physically and on the heterogeneous surface as multilayer adsorption. The Thales-based model demonstrated that after nine cycles of adsorption and formal regeneration with acid, the LDC adsorbent consistently maintained lead concentrations below the World Health Organization (WHO) standard.

1. Introduction

Employing waste-based adsorbents (WBAs) can be beneficial to reduce decontamination costs and ensure continuity between solid waste management and water treatment programs (Alswat et al., 2023). In recent years, WBAs have increasingly been used as a cost-effective and environmentally friendly method to remove hazardous substances from water resources, including heavy metals (Gkika et al., 2022; Varada et al., 2019). Various natural materials, such as agriculture waste (Bai et al., 2023), coir pith (Stelte et al., 2023), clay (Santoso et al., 2023), leaves (Alswat et al., 2023), and walnut shell (Enache et al.,

2023), have been used as green adsorbents due to their high adsorption capacity, low toxicity, and cost-effectiveness. In addition, waste-based inorganic substances such as zeolite, dolomite and bentonite can be used as adsorbents for heavy metals removal from water (Alyasi et al., 2025; Gruszecka-Kosowska et al., 2017; Kumari et al., 2025; Lee and Jo, 2010).

Generally, WBAs are categorized into three sources: industrial waste, construction and demolition waste (CDW), and agricultural waste (Zhou et al., 2022). CDW refers to waste from construction, renovation, and demolition, including concrete, brick, sand and gravel (Jain, 2021; Yuan et al., 2011). CDW is considered one of the most essential types of WBAs,

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as it has the potential to promote the circular economy and sustainable development goals (Sharma et al., 2021). Waste management strategies for used concrete emphasize immobilization and stabilization, where binders or additives encapsulate hazardous leachates and enhance durability performance. Recycling converts CDW into recycled aggregate concrete through closed-loop processes, while reuse focuses on design for deconstruction, enabling structural components to be reapplied across life cycles. Together, these strategies reduce landfill burdens, conserve natural aggregates, and optimize life-cycle sustainability (Xia et al., 2020). The considerable quantity of CDW generated, along with its potential deployment as a WBA, emphasizes its role as a practical solution for hazardous substance removal (Sharma et al., 2021).

One of the new technologies in the construction business is lightweight concrete, which has the benefits of structural engineering. However, when consumption rises, waste output also increases, especially for materials like autoclaved aerated concrete (AAC) (Gheibi et al., 2022). AAC is a popular lightweight building material typically consisting of silica, sand, cement, lime, quartz, and aluminum powder (Walczak et al., 2015). Due to their lighter weight, these materials are often used as substitutes for traditional bricks in non-load-bearing and partition walls (Aliabdo et al., 2014). Recent estimates place global annual non-reinforced AAC production at around 450 million m³ (Abhilasha et al., 2023). In 2020, Germany generated over 1 million m³ of post-demolition AAC, while the United Kingdom contributed around 700,000 m³. Eastern Europe exhibited even higher volumes, with Poland producing approximately 1.8 million m³ and Russia about 3.9 million m³ in the same year. Projections indicate that Europe's post-demolition AAC volumes will nearly double over the next decade, rising from 12.3 million m³ to 22.0 million m³ (Steins et al., 2022). To lessen the environmental burden of AAC waste, methods for its valorization should be developed in accordance with the principles of Integrated Solid Waste Management (ISWM) (Marshall and Farahbakhsh, 2013). Reuse of CDW is an approach that can be implemented in countries with high levels of re-construction activity. By adopting CDW management strategies, developing countries can address the challenges of waste management and promote sustainable and circular construction practices that align with the United Nations' Sustainable Development Goals (dos Reis et al., 2020). According to Lee et al. (2024), CDW can be effectively managed through several strategies, including reduction, reuse, recycling, incineration, and landfilling prioritizing the first three for environmental sustainability. Reduction focuses on optimized design and construction planning, while reutilization stresses selective deconstruction. Recycling repurposes concrete, plastics, wood, and metal wastes for construction materials or energy sources. Additionally, adsorption applications are highlighted, where certain CDW-derived materials, such as concrete fines or brick dust, are processed and used as low-cost adsorbents for removing pollutants from wastewater. These materials demonstrate significant potential for environmental remediation, offering a dual benefit of waste reduction and water purification. The appropriate method depends on material type, contamination level, and intended application.

Various methods are used for heavy metal removal from wastewater, including membrane, chemical, electric, and photocatalytic techniques. Among them, adsorption stands out for being cost-effective, technically mature, and environmentally friendly, though it has lower automation potential. It is widely favored for its simplicity, high efficiency, and ability to use low-cost adsorbents like industrial and construction wastes (Qasem et al., 2021).

Upon reviewing the literature, it is evident that various studies have been conducted on using CDW-based adsorbents to remove heavy metals from aquatic environments. For example, Coleman et al. (2005) investigated the efficacy of crushed concrete in removing three heavy metals (Zn²⁺, Cu²⁺, and Pb²⁺) from water. Djeribi and Hamdaoui (2008) also explored using mixed cedar sawdust with crushed bricks as an adsorbent to remove Cu²⁺ from water resources. In addition, Bibi et al. (2015) assessed the potential of cement as an adsorbent for arsenic from water.

Furthermore, Tsai et al. (2019) employed an innovative approach by using an aluminosilicate composite derived from waste display panel glasses as an adsorbent for removing Cu²⁺, Zn²⁺, and Ni²⁺ contaminants and investigating the underlying adsorption mechanism.

Praneeth et al. (2022) recently developed a novel cement-based mixture combined with biochar to remove several heavy metals. The study examined the interactions between different metals and evaluated the environmental protection potential of the adsorbent. Meanwhile, Yurk et al. (2022) investigated the effectiveness of concrete particles in removing Cu²⁺ and analyzed the hydromechanics of the process. In another study, Liu et al. (2022) produced a nano ecological recycled concrete (nano-ERC) with copper oxide nanoparticles to remove heavy metals, including As ions, Cd²⁺, Mn²⁺ and Pb²⁺ from simulated wastewater effluent. Recycled aggregates were used to reduce treatment costs and environmental burdens. Similarly, Sudagar et al. (2023) investigated low-grade Portuguese metakaolin, natural zeolite, and waste cork residue as replacements for commercial metakaolin-based geopolymers. The results showed that the applied waste-based polymers containing cork residue can effectively adsorb heavy metal cations such as Pb²⁺, Cd²⁺, Cu²⁺, and Zn²⁺ with a high level of selectivity. Masoumi and Ghaemi (2024) hypercrosslinked waste polycarbonate via Friedel–Crafts reaction to remove metallic ions from wastewater. In the investigation, the adsorption behavior of Pb²⁺ and Cd²⁺ ions was tested under varied conditions. Zennaki et al. (2023) investigated recycling waste-expanded polystyrene into anion exchange resins for Pb²⁺ and Cu²⁺ adsorption. Different modification techniques were examined to find the highest adsorption capacity and reusability. Radenković et al. (2024) explored using activated sunflower material (ASM) to remove Pb²⁺ and Cu²⁺ ions from wastewater. Optimal conditions were determined using Box-Behnken Design, with ASM showing high efficiency.

This study proposes reusing modified AAC waste, referred to as Low-Density Concrete (LDC), as an efficient adsorbent for removing Pb²⁺, Co²⁺, and Mn²⁺ from aquatic environments. LDC was characterized using X-Ray Fluorescence (XRF), Fourier Transform Infrared (FTIR), Brunauer-Emmett-Teller (BET), X-ray diffraction (XRD) analysis and Field Emission-Scanning Electron Microscopy (FE-SEM). Adsorption behavior was evaluated through isotherm modelling, adsorption kinetics using nonlinear Pseudo-First-Order (PFO) and Pseudo-Second-Order (PSO) models, and selectivity assessments under different concentration scenarios. The study further investigated competitive adsorption and morphological changes during the process. A Thales-based geometrical framework, the Adsorbent Regeneration Prediction Model (ARPM), is introduced for the first time to estimate regeneration cycles. The main objectives include implementing circular economy principles through construction waste reuse, demonstrating LDC characterization within Integrated Solid Waste Management (ISWM), determining adsorption mechanisms, and assessing regeneration potential. Key novelties are the simultaneous removal of three heavy metals using lightweight CDW, the development of ARPM to evaluate resiliency against pollution scenarios, and the integration of Sustainable Materials Management (SMM) with adsorption analysis.

It is important to note that the SMM concept has rarely been used to plan, track, and improve waste reduction with life-cycle-based metrics, as shown by Anshassi et al. (2019), or to support sustainable waste management through source reduction, material recovery, and energy recapture, as noted by Hall (2017). In this study, SMM is applied to select materials and assess the sustainability of an adsorbent. Also, the Thales' theorem-based model has not been used in this field before and is newly developed through this investigation. Qin et al. (2024) proposed simultaneous adsorption regeneration with sintered activated carbon based on kinetics modelling, and Yasri and Roberts (2024) outlined electrochemical regeneration concepts using mechanistic electrochemical modeling approach. However, their models remain physical and computationally complex for operators, while the ARPM introduced here simplifies calculations into a more practical and operator-friendly framework.

2. Materials and methods

The research roadmap of the present study is shown in Fig. 1. LDC waste was prepared and characterized by different tools in the first step. In the next step, the adsorption process of three heavy metals, including Pb^{2+} , Co^{2+} , and Mn^{2+} , was optimized according to pH, mass of adsorbent, and contact time. In the third step, the isothermal behavior of LDC in the interaction of Pb^{2+} , Co^{2+} , and Mn^{2+} was assessed based on eight different isotherm models, including Langmuir, Freundlich, Temkin, Dubinin-Radushkevich, Khan, Toth, Redlich-Peterson, and Sips. Furthermore, in the next phase of this study, the kinetics of heavy metal adsorption onto LDC was analyzed using PFO and PSO models. In addition, morphological changes were examined through elemental surface analysis before and after adsorption. The performance of LDC in heavy metal decontamination was also evaluated under competitive conditions to assess its selectivity and efficiency in multi-metal systems. Then, a Thales theorem-based model was presented to predict the regenerative adsorption cycle for each metal ion separately. Finally, the application of LDC waste material was assessed per the Sustainable Materials Management model.

2.1. Applied materials

A 1 g L^{-1} stock solution of Pb^{2+} , Co^{2+} , and Mn^{2+} was purchased from Merck, Germany, for the experiments. Additionally, in some tests, the solutions were prepared using Lead(II) acetate trihydrate (Sigma-Aldrich, USA), Cobalt(II) nitrate hexahydrate (Sigma-Aldrich, USA), and Manganese(II) sulfate monohydrate (Lach-Ner, Czech Republic). For pH adjustment of the solutions, HCl (0.1 mol L^{-1}) and NaOH (0.1 mol L^{-1}) from Merck, Germany are applied. Likewise, due to regeneration of the adsorbent, HNO_3 (1 mol L^{-1}), Merck, Germany was utilized.

2.2. LDC adsorbent preparation

The standard mixture consisted of Silica sand (350 kg m^{-3}), Water (305 kg m^{-3}), Cement Portland type II (25 kg m^{-3}), Lime (115 kg m^{-3}), and Aluminum powder (40 kg m^{-3}). The LDC mixture was produced at Razavi Block Company in Mashhad, Iran, by mixing and casting the raw materials to achieve a density of 500 kg m^{-3} . The mixture was autoclaved at $101,325 \text{ Pa}$ pressure and 150°C for 3 h. After synthesis, the LDC was washed three times, dried, and crushed. The resulting materials were sieved through mesh No. 140 (0.105 mm) (He et al., 2025; Kang et al., 2025). During the production process of LDC, chemical reactions generated gases, which created micropores, macropores, and interstices in the LDC structure. This significantly increased its porosity and surface area (Qiao et al., 2008).

2.3. Characterization

To determine the chemical composition of the LDC adsorbent, XRF

(Philips PW 1480, Netherlands) was utilized. FTIR (Thermo Nicolet model AVATAR 370, USA) was used for the exact characterization of the chemical specification, with a concentration on qualitative identification of various organic and organic/metallic molecules, determination of the molecular structure of different species, and identification of functional groups in the structure of the adsorbent. Additionally, in some secondary FTIR analyses, spectra with a resolution of 4 cm^{-1} were obtained using a germanium ATR crystal (NICOLET iZ10, Thermo Scientific, Waltham, MA, USA), equipped with a horizontal ATR accessory featuring a single reflection angle of 45° .

The morphological evaluation of the adsorbent surface was conducted using a FE-SEM model Philips S360, manufactured by Oxford in the United Kingdom. Prior to imaging, the adsorbent surface was prepared by coating it with a thin layer of gold using a sputter coater. The imaging was performed under vacuum conditions at an accelerating voltage of 10 kV . The resulting images were analyzed using appropriate software (point electronic GmbH control software) to obtain information about the surface morphology, such as pores, cracks, and irregularities. Additionally, in some other tests, EDS and SEM analyses were performed using a Tescan Vega XMU SEM (TESCAN s.r.o., Brno, Czech Republic), which is equipped with Energy-Dispersive X-ray Spectroscopy (EDS) and elemental mapping capabilities. The specific surface area of the adsorbent was measured using the gas adsorption method (BET instrument Belsorp mini II, Japan). The XRD analysis of LDC was performed using a Rigaku MiniFlex 600 diffractometer with $\text{CuK}\alpha$ radiation ($\lambda = 1.5406 \text{ \AA}$, operated at 40 kV and 30 mA), scanning from 10° to 70° with a step size of 0.01° per minute. Then, the results of the XRD analysis are evaluated using a program developed in MATLAB 2019b as part of this research.

2.4. Experimental practices and study of the effect of operational condition

The pH of a solution is a critical factor in removing ions from aqueous solutions. It affects the surface charge of the adsorbent and the degree of ionization and speciation of heavy metals in the solution (Yan et al., 2016). To prevent the precipitation of heavy metal ions, we studied the effect of pH on cation removal in the acidic pH range of 2.0 to 5.0, where the studied heavy metals are present in a cationic form (Harris, 2010).

The experimental set-up was a batch system to assess LDC performance in heavy metal adsorption. The fixed amount of sorbent (6.67 g L^{-1}) was added to 15 ml of metal solution at a concentration of 10 mg L^{-1} (with a dilution of 1 g L^{-1} stock solution of metal ions). The concentration was determined based on the study by Martemianov et al. (2017) and reflects levels that may be present in certain water sources, such as fish farms (Olaifa et al., 2003). The pH and temperature were fixed at 5.0 ± 0.1 (Metrohm 827 pH, Switzerland) and $25^\circ \text{C} \pm 0.1$, respectively. The effect of contact time was evaluated between 0–120 min with 400 rpm stirring using the one-factor-at-a-time (OFAT) method. Next, the optimal amount of adsorbent (LDC) was examined in the 0.02–0.15 g range.

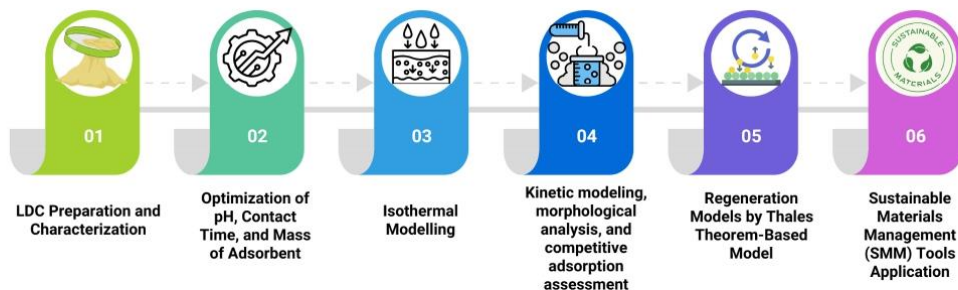


Fig. 1. The research roadmap of the present study.

After a selected contact time, separation was performed by centrifuging at 3000 rpm for 10 min (Andreas Hettich D72, Tuttlingen, Germany). For heavy metal pollution detection, AAS was used (Perkin Elmer AAnalyst 700 Atomic Absorption Spectroscopy with specific lamps for each metal Pb: 283.3 nm, Co: 240.7 nm, and Mn: 279.5 nm) according to Farukh (2012) and Loon (2012). Furthermore, in the kinetics and competitive adsorption tests, heavy metal concentrations were measured using ICP-OES (Thermo Scientific, USA).

2.5. Adsorption isotherms and regeneration cycles

Two-parameter isothermal models, including Freundlich and Langmuir, were initially employed to evaluate the isothermal behavior of LDC for each heavy metal. Langmuir is a monolayer adsorption isotherm that limits the adsorption to a single molecular layer. According to this model, increasing the distance from the adsorbent surface significantly reduces the intermolecular forces (Foo and Hameed, 2010). Freundlich adsorption isotherm is an empirical equation and one of the most practical approaches to analyzing adsorption (Foo and Hameed, 2010). This method is more suitable for evaluating adsorption on heterogeneous surfaces, mainly when dealing with low to moderate concentrations. However, due to the close coefficient of determination in the mentioned models, three-parameter techniques, such as Khan, Toth, Redlich-Peterson, and Sips, were utilized. Finally, a combination of these models was used to determine the adsorption mechanism for each heavy metal. All equations and models applied in this part of the research are summarized in Table S1. Comparably, two-parameter models such as the Temkin and Dubinin-Radushkevich isotherms are used to accurately determine the principles of adsorption in a variety of physical, intra-particle diffusion, or chemical classifications (Gheibi et al., 2022).

In this study, the regenerability of the adsorbent is evaluated over five cycles. During each adsorption-desorption cycle, 1 mol L⁻¹ HNO₃ is used to regenerate heavy metal ions from the adsorbent surface. The regeneration performance is assessed based on the optimal interaction of the adsorbent with the heavy metal ions and in industrial concentration of the heavy metals.

2.6. Kinetics analysis

In the present study, kinetic analyses were conducted under controlled conditions: pH = 5 and adsorbent dosage = 3.34 g L⁻¹. The reaction was monitored over a time range of 0–120 min, with a fixed reactor volume of 30 mL. Also, the initial concentration of heavy metals in the solution is adjusted around 5 mg L⁻¹. The experimental data were evaluated using two kinetic models: pseudo-first-order (PFO) and pseudo-second-order (PSO) (Table S2) (Wang and Guo, 2020). The performance of each model was assessed based on the coefficient of determination (*R*²) and the differences between the experimental and theoretical adsorption capacities.

2.7. Morphology changes analysis

To better understand the reaction mechanism, morphological changes in the LDC surface were evaluated before and after adsorption using SEM and EDS characterization. The objective of this analysis was to detect the presence of Mn²⁺, Co²⁺, and Pb²⁺ on the LDC surface. The adsorption experiments were conducted under controlled conditions: pH = 5, contact time = 30 min, adsorbent dosage = 3.34 g L⁻¹, solution volume = 30 mL, and metal ion concentrations of approximately 5 mg L⁻¹.

2.8. Competitive adsorption

To further understand the selectivity of the adsorbent, competitive adsorption tests among Pb²⁺, Co²⁺, and Mn²⁺ were conducted. The

experiments were performed under controlled conditions: pH = 5, contact time = 30 minutes, adsorbent dosage = 3.34 g L⁻¹, and solution volume = 30 mL. The competitive behavior was evaluated across three concentration scenarios: low, medium, and high. In each scenario, the metal ion with the highest affinity toward LDC was considered the primary adsorbate, while the other two acted as interfering ions.

2.9. Adsorbent regeneration prediction model (ARPM)

This research introduces the ARPM based on Thales' Theorem (Grecu et al., 2020), for predicting adsorbent performance in various cycles of water and wastewater treatment processes. In this new method, the trend of removal percentage in repeated adsorption cycles is evaluated with the geometrical Thales Theorem. Consideration of effective parameters including the number of cycles (*N*), initial concentration (*C*₀), remaining concentration in first adsorption cycle (*C*_{R1}), remaining concentration in last adsorption cycle (*C*_{Rn}) and allowable standard concentration of the metal ion in water resources (*C*_s), the number of cycle reaction that can meet standard thresholds are estimated. According to Fig. 2, reducing heavy metal removal percentage in different cycles is considered as a triangle, and Thales geometrical rules (Eq. (1)) are applied to it. In the mentioned equation Δ_i expresses the difference between the removal percentage in the first cycle and the removal percentage in the standard concentration threshold (considering allowable standard concentration) and, Δ_n express the difference of removal percentage in first and last cycles.

$$\frac{\Delta_i}{\Delta_n} = \frac{x}{N} \quad (1)$$

After simplifying Eq. (1) with fundamental definition of the parameters, the final mathematical model for determining the LDC regeneration cycle (*N*-x) is expressed in Eq. (2). In other words, after calculating the x-factor, the adsorbent should be regenerated to meet the standard of heavy metals in water resources.

$$x = N \times \frac{C_{Rn} - C_s}{C_{R1} - C_s} \rightarrow N - x = N \times \left(1 - \frac{C_{Rn} - C_s}{C_{R1} - C_s} \right) \quad (2)$$

The application of this model is valid only when the standard concentration lies within the range of concentrations being evaluated. In other words, the initial concentration must progressively decrease to meet the standard limits of the receiving water body over successive adsorption cycles. In this type of equation, the linearity condition of efficiency reduction across different adsorption cycles is first satisfied, followed by the triangulation required for the application of Thales' theorem.

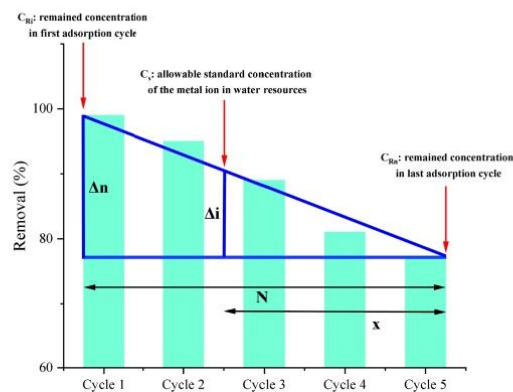


Fig. 2. Scheme of Thales-based ARPM in this research.

Through mathematical modeling, it becomes possible to determine the regeneration cycle of LDC adsorbent when dealing with unknown water or wastewater samples. In this study, the regeneration cycle is illustrated using the influent heavy metal content (Pb^{2+} , Co^{2+} , and Mn^{2+}) data from a wastewater treatment plant located in the Toos Industrial Park of Mashhad City, Iran. Additionally, Cs values conforming to WHO standards in river water are incorporated into the formula (Rozana et al., 2020; Waghmare et al., 2015). However, it is essential to note that regeneration tests are conducted individually for each metal ion under optimal conditions for heavy metals.

2.10. Sustainable materials management models

In this section of study, the applied concrete-based material as an adsorbent is analyzed based on Sustainable Materials Management (SMM) prioritization tools which was introduced by the Environmental Protection Agency (EPA), USA (US EPA, 2020). The lifecycle-based Sustainable Materials Management Prioritization Tools provide a basis for spotting chances to improve environmental sustainability in the creation and consumption of goods and services. They identify critical

environmental issues and hotspots, which helps with action prioritization and efficient resource allocation. More thorough information should be added to these tools to make exact decisions prior to taking action (Chen et al., 2017).

EPA's sustainability assessment uses a life-cycle-based analytic framework incorporating tools like Comprehensive Environmental Data Archive (CEDA 3.0) and U.S. Bureau of Economic Analysis 1998 input/output (BEA I/O) tables to evaluate 480 materials, products, and services across 17 criteria, including environmental impacts (e.g., toxicity, GWP), resource use, and waste. Impacts are assessed from direct, intermediate, and final consumption perspectives. A vector analysis method is applied to compute relative rankings and guide sustainability prioritization strategies (US EPA, 2015). The SMM prioritization framework applies life-cycle input-output (LCA-IO) modeling where economic activity vectors are multiplied by environmental extension coefficients. Using the Leontief inverse $(I - A)^{-1}$, total upstream requirements are derived. Indicators span environmental, resource, waste, and socio-economic. Computation involves normalizing impacts across categories and applying vector magnitude analysis to generate comparative sustainability scores (US EPA, 2015).

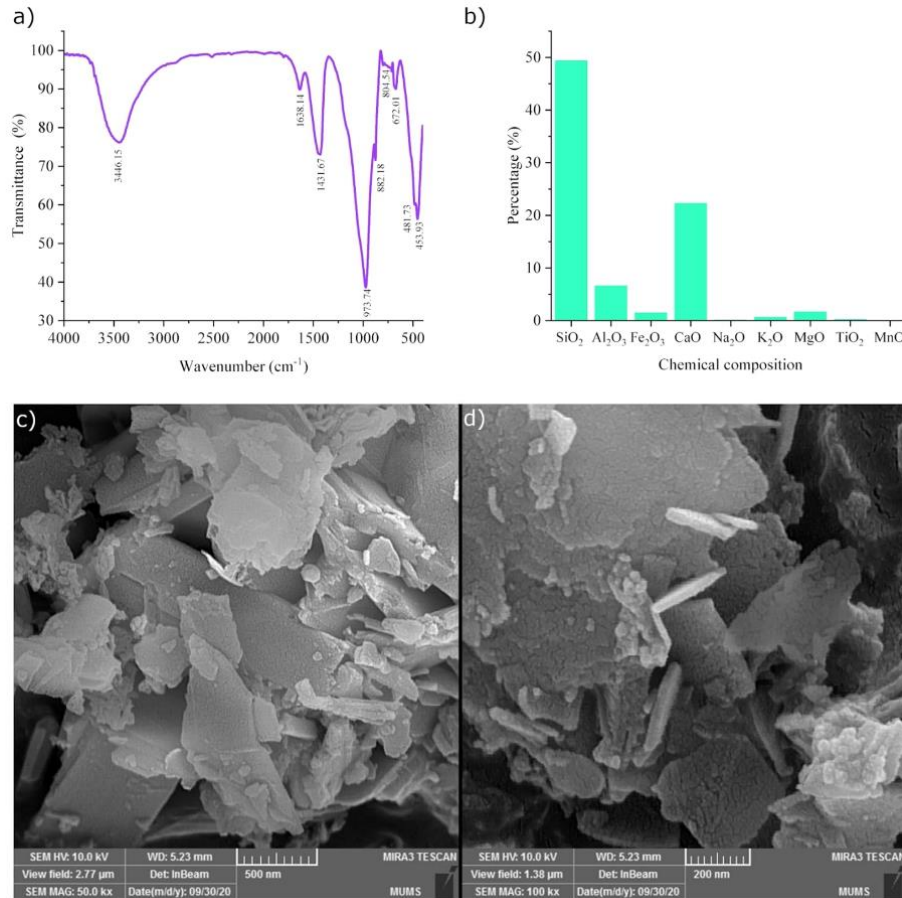


Fig. 3. The outputs of LDC adsorbent characterization based on (a) FT-IR spectrum, (b) XRF analysis, (c) FE-SEM, scale bar: 500 nm, and (d) FE-SEM, scale bar: 200 nm images.

The present study applied a concrete-based material for the decontamination of heavy metals (Pb^{2+} , Co^{2+} , and Mn^{2+}). While the application of carbon and graphite materials (such as Graphene Oxide-GO) are common for the decontamination of heavy metals from water resources (Li et al., 2015; Liu et al., 2011; Yan et al., 2014). Therefore, in this part of study, the sustainability indexes of both (1) concrete-based and (2) carbon/graphite-based materials are analyzed. The SMM analysis is done in five different categorizations summarized in Table S3 (US EPA, 2020).

3. Results and discussions

3.1. Characterization of adsorbent

The FTIR spectrum characterization is presented in Fig. 3a. The symmetric stretching of Si-O can be detected at 973 cm^{-1} . In addition, a weaker feature was observed around 672 cm^{-1} . The Si-O rocking vibration band can also be observed at 453 cm^{-1} , based on the FTIR results (Herth et al., 2016; Liang et al., 2012). CO_3^{2-} bands are present at 882 cm^{-1} (out-of-plane bend), and the asymmetric stretching signal of calcite/aragonite is demonstrated at 1431 cm^{-1} . The broad band at 3446 cm^{-1} is primarily related to O-H stretching vibration due to adsorbed water in the sample (Bergren et al., 1978).

To characterize the chemical composition of the LDC adsorbent, X-ray fluorescence (XRF) analysis was performed on the LDC samples (Fig. 3b). There were six oxides determined, with a majority of Al_2O_3 and CaO . The sum of measured chemical composition is not equal to 100 %, which indicates the presence of adsorbed water and carbonate compounds (Beckhoff et al., 2007). FE-SEM images of the LDC adsorbent (Fig. 3c,d) shows a high porosity that might cause a high affinity for the adsorption of heavy metals. Numerous investigations have demonstrated that adding aluminum powder to concrete increases its porosity (Van et al., 2019). Hydrogen gas is produced by the reaction between the aluminum powder and the calcium hydroxide in the concrete mixture as it cures. Additionally, during adsorption, this reaction increases the specific surface area and adsorption process efficiency. The specific surface area of LDC was determined to be $30.2\text{ m}^2\text{ g}^{-1}$, based on the N_2 adsorption isotherms measured by BET analysis. This is in a range of various CDW materials that are applied for adsorption process of contaminations, e.g., calcinated AAC (Gheibi et al., 2022), lightweight cellular concrete (Azizi et al., 2023), and ultra-lightweight concrete (Yu et al., 2015) have the surface areas of $24.64\text{ m}^2\text{ g}^{-1}$, $12.86\text{ m}^2\text{ g}^{-1}$, and $50\text{ m}^2\text{ g}^{-1}$, respectively.

3.2. Study the effect of pH on adsorption

The result of studying the effect of pH on adsorption is shown in Fig. 4a. The poor adsorption at low pH is due to the protonation of adsorption sites, resulting in a net positive charge on the surface (Xu, 2013). The observed difference in the removal rate of different metal

ions at low pH can be attributed to the varying binding between the adsorbent functional groups and metal ions, depending on the charges of the metal ions and the specific characteristics of the adsorbent surface (Mathialagan and Viraraghavan, 2002). As it is clear from Fig. 4a, the maximum removal (%) can be seen at the maximal pH considered in the experiment, $\text{pH}=5.0$, for all three ions.

Based on the solubility product constant (K_{sp}) in the dissolution of metal ions in water samples, precipitation can occur for Pb^{2+} ($K_{sp}=8 \times 10^{-16}$), Co^{2+} ($K_{sp}=1.3 \times 10^{-15}$), and Mn^{2+} ($K_{sp}=1.6 \times 10^{-13}$) (Harris, 2010) at pH levels of ~ 7.5 , 7.79 , and 8.82 respectively, with a concentration of 200 mg L^{-1} of heavy metals in the reactor. 200 mg L^{-1} was the maximum concentration of heavy metals which is applied and therefore, for other concentrations, the pH of precipitation will be higher than the above-written values (Harris, 2010). Thus, this study aims to evaluate the pure adsorption process for all selected metal ions within the same environment, thus, pH 5 can be considered the ultimate value in the assessment.

The chemistry of the heavy metal ions and the metal oxide adsorbent materials (such as SiO_2 , CaO , and Al_2O_3) can both explain the phenomenon, which indicates that the removal percentage of heavy metals in adsorption processes is lower at more acidic pH ($\text{pH}=2$). The major metal oxides present have a net positive charge on their surfaces at low pH. For example, SiO_2 's surface tends to be positively charged at low pH levels because protonated silanol groups (Si-OH^{2+}) are present (Brown et al., 2014; Ho and McKay, 1999; Lagergren, 1907).

Pb^{2+} showed higher adsorption at pH 2 than Co^{2+} , and Co^{2+} higher than Mn^{2+} . This is because Pb^{2+} attaches to the adsorbent surface more readily than Co^{2+} and Mn^{2+} because of their more positive charge (Xu, 2013) and could make surface complexes more easily than Co^{2+} or Mn^{2+} (Dzombak and Morel, 1991; Sparks et al., 2022; Stumm and Morgan, 2013).

3.3. Optimum contact time and doses of LDC

As shown in Fig. 4b, it can be understood that the optimum contact time for both Co^{2+} and Pb^{2+} is 20 min. After this time, the Removal Percentage (RP) for heavy metals reached 98 % for Pb^{2+} and 80 % for Co^{2+} . During the 120-minute kinetic experiments, there was only a negligible change in RP, around 1 % for Pb^{2+} and around 4 % for Co^{2+} . Therefore, 20 min can be considered the optimal contact time for the adsorption of these ions, whereas for Mn^{2+} , 30 min is identified as the optimal reaction time.

The behavior of adsorbents toward heavy metals varies. However, in this study, the minimal contact time to get equilibrium adsorption was found to be between 20 and 30 min (Co^{2+} and Pb^{2+} : 20 min, Mn^{2+} : 30 min). Panda et al. (2020) demonstrated that significant removal of metal ions (over 80 %) occurred in 0–20 min of contact time when various heavy metals were adsorbed onto geopolymer made from dolochar ash. The fastest adsorption in the first phase of the test is attributed to the presence of numerous binding sites on the surface of dolochar ash

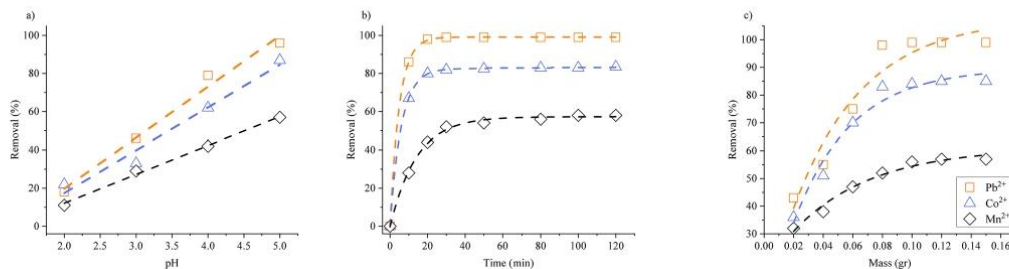


Fig. 4. Heavy metals removal as a function of (a) pH, (b) time, and (c) LDC dosage, general conditions: 10 mg L^{-1} of each heavy metal, $\text{pH}=5$, 6.67 g L^{-1} of LDC powder, 30 min contact time. Repeatability error for Pb^{2+} : less than 1%, Mn^{2+} : 7%, and Co^{2+} : 4.3%.

geopolymer. Similar trends in adsorption efficiency have been observed (Siyal et al., 2016), where saturation of adsorption of heavy metal ions on potent adsorbents like fly ash, coal, and geopolymeric Linz-Donawitz (LD) slag typically occurs within 30–60 min (Elimbi et al., 2011; Sarkar et al., 2017). Senberber et al. (2017) demonstrated that in the application of borax sludge for decontamination of chromium ions, maximum performance can be achieved in less than 30 min. Likewise, Bulut and Tez (2007) proved that in the adsorption of heavy metals by sawdust as a waste material, maximum adsorption can be observed in less than 30 min of the reaction.

The adsorption mechanism of Pb^{2+} and Zn^{2+} using slaked lime ($\text{Ca}(\text{OH})_2$) is based on the precipitation of metal hydroxides. Lime dissolves to form Ca^{2+} and 2OH^- ions, which react with metal ions in water. Pb^{2+} and Zn^{2+} form $\text{Pb}(\text{OH})_2$ and $\text{Zn}(\text{OH})_2$, respectively, as precipitates. The efficiency depends on maintaining an optimal pH range. High alkalinity from lime can cause re-dissolution of amphoteric hydroxides, reducing removal efficiency (Wang et al., 2016). Harja et al. (2017) demonstrated that the adsorption of Pb^{2+} onto lime-based adsorbent reached its maximum within 50–60 min. Consequently, the evaluated LDC exhibited synergistic effects, including the influence of the CaO compound, which enhanced the kinetic behavior. Pb^{2+} sorption on $\gamma\text{-Al}_2\text{O}_3$ involves a rapid initial adsorption phase followed by a slower phase, consistent with an inner-sphere bidentate bonding mechanism. The rapid phase, which accounts for 76 % of total Pb^{2+} adsorption within 15 min, involves Pb binding to surface aluminol groups (Strawn et al., 1998). Therefore, in the present study, the rapid adsorption of heavy metals can be attributed to the presence of aluminum powder in the adsorbent mixture. In another study, Tabesh et al. (2018) demonstrated that the adsorption of Pb^{2+} and Cd^{2+} onto $\gamma\text{-Al}_2\text{O}_3$ nanoparticles resulted in the elimination of over 80 % of the contaminants within the first 10 min. In contrast, in the present study, around 5 % of the concrete weight consists of aluminum oxide, which provides an additional driving force in the LDC reaction, enhancing the kinetic performance of the adsorption process.

According to Fig. 4c, it is evident that an adsorbent amount of 6.67 g L^{-1} in the reactors yields sufficient results. In the case of Pb^{2+} , the RP value remains unchanged between 0.1 g and 0.15 g ($6.67\text{--}10 \text{ g L}^{-1}$), and for Co^{2+} and Mn^{2+} , the changes were only 1.17 % and 1.75 %, respectively. Therefore, 0.1 g (6.67 g L^{-1}) of LDC mass versus RP is sufficient, indicating its optimal value.

In the present investigation, the choice of 0.105 mm particle size was made to highlight the effect of size on adsorption performance. Smaller particles generally enhance adsorption due to their higher surface area and shorter diffusion pathways, which accelerate ion transfer and increase active site accessibility (Suresh Kumar et al., 2019). However, very fine particles often lead to suspension stability issues and hinder solid-liquid separation. Conversely, larger particles settle easily but reduce adsorption kinetics because of limited surface exposure (Sharma, 2018). The selected size provided a balance between high adsorption efficiency and practical handling, ensuring good reactivity while avoiding excessive colloidal behavior.

3.4. Adsorption studies

The experimental data determined for all three heavy metals were fitted with all types of considered isotherms (two and three parameters) with the aim of understanding the mechanisms of the adsorption process.

3.4.1. Two parameter isotherms

3.4.1.1. Langmuir isotherm. The data can be easily fit with the Langmuir adsorption isotherm with a high R^2 equal to 0.97, 0.93 and 0.89 for Pb^{2+} , Co^{2+} and Mn^{2+} adsorption, respectively (Table 1). Therefore, the correlation of the Langmuir isotherm for Pb^{2+} is greater than for the

Table 1
Results of the study of two-parameter isotherms.

Two Parameter Isotherm	Parameter	Co^{2+}	Pb^{2+}	Mn^{2+}
Langmuir	Q_{max} (mg g^{-1})	15.17	43.1	23.52
	K_{ads} (L mg^{-1})	0.0326	2.86	0.0115
	R^2	0.93	0.97	0.89
Freundlich	$1/n$	0.525	0.703	1.01
	K_F (mg g^{-1})	1.05	40.43	0.32
	R^2	0.98	0.99	0.99
Temkin	K_T (L mg^{-1})	0.82	77.3	0.26
	b (J mol^{-1})	1027.53	389.9	675.7
	R^2	0.88	0.88	0.96
D-R	Q_m (mg g^{-1})	7.88	24.92	8.54
	K ($\text{mol}^2 \text{kJ}^{-2}$)	$1\text{E-}6$	$2\text{E-}8$	$8\text{E-}6$
	E (J mol^{-1})	707.1	5000	250
	R^2	0.82	0.96	0.89

others. The Langmuir model assumes that adsorption occurs in a monolayer and only at specific, fixed sites that are identical in energy and equivalent (Foo and Hameed, 2010). There are no interactions or hindrances between adjacent molecules on these sites. Furthermore, Langmuir's theory associates a rapid decrease in intermolecular attractive forces with increasing distance (Foo and Hameed, 2010). Therefore, the Langmuir isotherm assumes uniform adsorption, where each molecule has constant enthalpies and sorption activation energy, and all sites have equal affinity for the adsorbate. Additionally, there is no movement of the adsorbate across the surface (Radenković et al., 2024). Graphically, Langmuir adsorption model is represented by a curve with a plateau, indicating saturation where increasing solute concentration does not cause further adsorption because all adsorption sites are occupied.

When comparing the heavy metal, it should be mentioned that Langmuir adsorption capacity (Q_{max}) is highest for Pb^{2+} (43.1 mg g^{-1}) > Mn^{2+} (23.52 mg g^{-1}) > Co^{2+} (15.17 mg g^{-1}). Therefore, the strength of adsorption of Pb^{2+} with LDC was more than that of Mn^{2+} and Co^{2+} . In this study, the hydrated radius of Pb^{2+} ($4.01 \text{ \AA}$) is smaller than that of Mn^{2+} ($4.38 \text{ \AA}$) and Co^{2+} ($4.23 \text{ \AA}$). Therefore, the hydration of Pb^{2+} by water molecules will be lower compared to the other two ions. Consequently, the unoccupied active sites of Pb^{2+} can participate more readily in adsorption stages with LDC compared to Mn^{2+} and Co^{2+} . This could be a contributing factor to the higher adsorption capacity of Pb^{2+} compared to Mn^{2+} and Co^{2+} (Goel et al., 2004).

A dimensionless constant, referred to as the separation factor (R_L) defined by Webber and Chakravorti, can be expressed as Eq. (3) (Weber and Chakravorti, 1974).

$$R_L = \frac{1}{K_{\text{ads}} C_0 + 1} \quad (3)$$

where K_{ads} and C_0 represent the Langmuir constant (L mg^{-1}) and initial concentration of contamination (mg L^{-1}), respectively. The computations demonstrated that the R_L for Pb^{2+} for all concentrations between $10\text{--}200 \text{ mg L}^{-1}$ has a value between 0.03 to 0.001 values. Therefore, the $0 < R_L < 1$ and it is a favorable adsorption process. Also, the R_L has value between 0.3 to 0.9 for the same concentrations of Mn^{2+} . While in 10 mg L^{-1} concentration of Mn^{2+} , the R_L is near to 1 and it means that at a lower initial concentrations, the adsorption process will be done linearly ($R_L=1$). Finally, in the case of Co^{2+} , the $0.1 < R_L < 0.75$ and therefore, the process of adsorption is favorable like to Pb^{2+} one (Foo and Hameed, 2010). The curve-fitting results of the Langmuir isotherm model for all applied metal ions are represented in Fig. S1a.

3.4.1.2. Freundlich isotherm. The second isotherm tested was Freundlich. The Freundlich isotherm suggests multilayer adsorption on heterogeneous surfaces. It indicates that adsorption capacity increases as concentration rises, with multiple adsorption layers forming at different energy levels (Foo and Hameed, 2010; Ramadan and El-Masry, 2024).

Assessing whether the measured data conforms to the Freundlich isotherm can best be done by displaying them on a log-log scale. While the data for Co^{2+} can be fitted by a straight line, the data for the other two metals show a decrease in the adsorbed amount for the highest concentration in solution. This shows saturation of sorption and therefore, in the case of Pb^{2+} and Mn^{2+} the data were plotted as a straight line without the point corresponding to the highest concentration. Then the best fit of all applied metallic ions to the Freundlich model was over 0.98 (Table 1) and the Freundlich isotherm is valid up to 120 mg L^{-1} for Co^{2+} , 40 mg L^{-1} for Mn^{2+} , and 0.2 mg L^{-1} for Pb^{2+} .

The analysis of the model confirmed that the adsorption processes of Pb^{2+} , Co^{2+} , and Mn^{2+} onto LCD wastes occur with multilayer adsorption and on heterogeneous surfaces. However, the results of Pb^{2+} adsorption were fitted to both Langmuir and Redlich-Peterson, and Sips). In total, the Freundlich constant of Pb^{2+} equal to 40.43 mg g^{-1} was considerably higher than both Mn^{2+} and Co^{2+} . Consequently, the flux of the lead adsorption process exceeded that of the other two metals in the mass transfer process of adsorption. Additionally, in the lead adsorption process, the $1/n$ value was lower than that of the other two metals, indicating cooperative procedure (Haghseresht and Lu, 1996) in the lead adsorption onto LDC waste compared to Co^{2+} and Mn^{2+} . The results of curve fitting in the Freundlich model are demonstrated in Fig. S1b.

3.4.1.3. Temkin isotherm. According to Table 1 and Fig. S1c, it is evident that the adsorption of Mn^{2+} ions onto LDC wastes followed the Temkin isotherm model more closely than Pb^{2+} and Co^{2+} . The R^2 coefficient for Mn^{2+} was ~ 0.96 , while for both Pb^{2+} and Co^{2+} it was ~ 0.88 . The Temkin isotherm includes a factor that explicitly considers adsorbent-adsorbate interactions. By disregarding extremely low and high concentrations, the model assumes that the heat of adsorption (which is dependent on temperature) of all molecules in the layer would decrease linearly rather than logarithmically with coverage. As shown in the equation (Table S1), its derivation assumes a uniform distribution of binding energies, up to a certain maximum binding energy (Kim et al., 2004). Consideration to Table 1, it is clear that b value for all ions was less than 8 kJ mol^{-1} and therefore, the adsorption of Pb^{2+} , Co^{2+} , and Mn^{2+} were carried out physically onto LDC wastes. However, the certainty of this result for Mn^{2+} is greater than two others because of its coefficient of determination values, which will be evaluated more in D-R isotherm assessment.

3.4.1.4. Dubinin-Radushkevich (D-R) isotherm. D-R isotherm parameters for each metal are given in Table 1 and Fig. S1d. Considering $R^2 > 0.96$, only Pb^{2+} follows the D-R isotherm. Besides, all values obtained for parameter E from Table 1 are less than 8 kJ mol^{-1} (Demiral and Güngör, 2016), which indicates the physical nature of the adsorption process for all metals. Results obtained from the E parameter verify all conclusions received from the b parameter in the Temkin isotherm model about the nature of the adsorption, which is physical.

3.4.2. Three parameter isotherms

All the R^2 values for studied heavy metals according to 3-parameter isotherms are higher than 0.94, indicating the high versatility of heavy metals adsorption onto LDC adsorbent according to this model.

3.4.2.1. Khan isotherm model. The Khan isotherm is a generalized model proposed for pure solutions, where a_k and b_k represent the model exponent and model constant, respectively. With relatively high correlation coefficients and minimum sum square error or chi-square values, it is possible to accurately determine its maximum uptake values (Khan et al., 1997). The coefficient of determination of Khan isotherm for Co^{2+} , Pb^{2+} , and Mn^{2+} is more than 0.98 (Table 2 and Fig. S1e). However, it

Table 2
Results of the study of three-parameter isotherms.

Three Parameter Isotherm	Parameter	Co^{2+}	Pb^{2+}	Mn^{2+}
Khan	q_s	5168.53	15.42	11,203.43
	b_k	5.63E-5	8.25	2.67E-5
	a_k	154.71	0.59	318.29
	R^2	0.98	0.99	0.99
Toth	K_{Toth}	0.43	6.98	0.83
	a_T	0.13	0.01	0.77
	t	1.95	2.49	2.29
	R^2	0.96	0.99	0.94
Sips	K_S	73.88	0.06	0.05
	a_s	1.2	0.004	0.004
	β_s	0.85	1.6	1.6
	R^2	0.97	0.99	0.99
R-P	K_R	0.22	73.13	0.23
	a_R	9.17E-6	30.72	2.24E-7
	g	2.46	12.81	3.24
	R^2	0.98	0.99	0.99

should be noted that a_k value does not converge to 1 for each metallic ion adsorption onto LDC waste, indicating that the adsorption process is not Langmuir. Consequently, it can be understood that with the application of a three-parameter adsorption process, the heterogeneous surface of the reaction is evident. In the following section, the accuracy of this claim will be analyzed using three other parameter isothermal models.

3.4.2.2. Toth isotherm model. The Toth isotherm model is an additional empirical equation designed to improve Langmuir isotherm fittings using experimental data. It is particularly effective in describing heterogeneous adsorption systems, as it satisfies both low and high-end concentration boundaries. This model assumes an asymmetric quasi-Gaussian energy distribution, where most sites have adsorption energies lower than the peak (maximum) or mean value (Ho et al., 2002). According to Table 2 and Fig. S1f, it is evident that the adsorption of Pb^{2+} , Mn^{2+} , and Co^{2+} follows the Toth model with a R^2 of more than ~ 0.94 . However, upon calculation, it becomes clear that the $1/t$ values for all metallic ions are around 0.5, significantly different from 1. Therefore, this three-parameter isothermal model supports Khan's assertion, confirming the heterogeneous surface of adsorption onto LDC wastes once again.

3.4.2.3. Sips isotherm model. The values of the Sips isotherm parameters for this study are presented in Table 2. According to the model, it can be understood that all metallic ion adsorption processes onto LDC waste have been fitted to Sips model with at least 0.97 correlation coefficient. Based on the B_s parameter values in Table 2 and the curve fitting results shown in Fig. S1g, it can be observed that under Sips isotherm conditions, B_s does not converge to 1. Consequently, it does not follow the Langmuir isotherm, indicating that the adsorption process for Pb^{2+} , Mn^{2+} , and Co^{2+} occurred on a heterogeneous surface, consistent with the findings of the Khan and Toth model.

3.4.2.4. Redlich-Peterson (R-P) isotherm model. The R-P model values for this research are listed in Table 2 and Fig. S1h. The values of g parameter for Pb^{2+} ($R^2 = 0.99$), Co^{2+} ($R^2 = 0.98$) and Mn^{2+} ($R^2 = 0.99$) metals are not close to 1 and therefore, the model's outcomes proved the heterogeneous surface of reaction. Finally, all two and three parameter isothermal equations determined the multilayer adsorption of the ions onto the adsorbent.

As a summary, various isotherm models were applied to fit the adsorption data in this study. The results from two-parameter models (Langmuir and Freundlich) and three-parameter models (Khan, Toth, Sips, and Redlich-Peterson) indicate that the adsorption occurs in multiple layers and the LDC surface is heterogeneous. Additionally, the Temkin and Dubinin-Radushkevich models prove that the adsorption of

Pb²⁺, Co²⁺, and Mn²⁺ onto LDC is dominated by physisorption.

According to Al-Maliky et al. (2021), concrete-based adsorbents possess a high point of zero charge (PZC) of ~12, meaning their surfaces are positively charged below this value and negatively charged above it, which dictates the adsorption pathway: favoring anions under acidic conditions and cations under alkaline ones. However, in LDC tested at pH 2–5, far below the ZPC, heavy metals such as Pb²⁺, Mn²⁺, and Co²⁺ were still efficiently removed, not by electrostatic attraction but through ion exchange with Ca²⁺ and Al³⁺ and complexation with surface hydroxyls. Adsorption capacities followed the trend Pb²⁺ (43.1 mg/g) > Mn²⁺ (23.5 mg/g) > Co²⁺ (15.2 mg/g), consistent with ionic size and binding affinity, with Pb²⁺ showing the strongest uptake. Thus, while concrete's surface charge condition governs adsorption behavior, LDC demonstrates that below-ZPC environments still enable effective cation removal through chemical interactions, emphasizing its sustainable potential in heavy metal remediation.

3.5. Kinetics analysis of heavy metal adsorption

The nonlinear kinetic modeling (Ait Ichou et al., 2023; Canzano et al., 2012) outcomes (Table 3 and Figs. S2–S4) clearly indicate that the PSO model provides a superior description of adsorption kinetics compared to the PFO model for all three metal ions. This indicates that the adsorption rate may be influenced by the concentration of both the metal ions and the available active sites on the adsorbent surface (Wang and Guo, 2020). For Pb²⁺, the PFO fit yielded an R² of 0.69, RMSE of 0.065, and MAE of 0.05, while the PSO fit improved drastically with R² = 0.97, RMSE = 0.019, and MAE = 0.017. The theoretical q_e predicted by the PSO model (1.28 mg g⁻¹) was also closer to the experimental value than that of the PFO model (1.26 mg g⁻¹), suggesting that Pb²⁺ adsorption onto LDC is best explained by surface-controlled adsorption pathways. For Co²⁺, although PFO achieved a relatively high R² of 0.89, the PSO model again showed better agreement (R² = 0.90) while reducing MAE from 0.11 to 0.10 and improving q_e prediction from 1.55 mg g⁻¹ (PFO) to 1.72 mg g⁻¹ (PSO). Mn²⁺ adsorption followed the same pattern, with the PFO model giving R² = 0.51, RMSE = 0.15, and MAE = 0.11, whereas the PSO model improved these metrics to R² = 0.76, RMSE = 0.10, and MAE = 0.08, with q_e = 1.07 mg g⁻¹. Collectively, these numerical values demonstrate that PSO consistently provides a better statistical and predictive fit than PFO.

The figures provide analytical support for these statistical results. In Fig. S2 (Pb²⁺), the kinetic model comparison shows that the PSO curve overlaps almost entirely with the experimental data, whereas PFO underestimates adsorption at equilibrium. The residuals plot shows PSO residuals distributed narrowly within ±0.02 mg g⁻¹, compared to PFO which shows deviations up to 0.15 mg g⁻¹. The parity plot shows PSO points clustering tightly along the y = x line, while PFO deviates, especially at higher q_e values. Similarly, in Fig. S3 (Co²⁺) and Fig. S4 (Mn²⁺), the PFO model exhibits greater fluctuations compared to the

PSO model, as reflected by the wider distribution of residual values. These numerical reductions in residual ranges confirm that PSO reduces both random and systematic deviations from experimental kinetics.

These improvements quantitatively demonstrate that the adsorption kinetics follow a PSO mechanism, indicative surface complexation dominated by electron exchange or sharing. Pb²⁺ exhibits the most pronounced strong surface binding behavior with near-perfect PSO fitting, Co²⁺ shows moderate but consistent PSO superiority, and Mn²⁺ presents weaker but still statistically significant improvement. This tiered performance suggests different binding strengths and mechanisms among the ions, with Pb²⁺ having the strongest affinity toward LDC surfaces.

The result suggests a stronger interaction or faster uptake of Pb²⁺ by the adsorbent, potentially due to its physicochemical properties such as hydration energy or ionic radius (Conway and Ayranci, 1999). Fig. S2–S4 illustrates this trend through the steeper slope observed for Pb²⁺ uptake over time. As a result, the findings highlight that the PSO model provides a more accurate and reliable description of the adsorption kinetics for all three metal ions, with Pb²⁺ exhibiting the highest rate constant and most rapid adsorption behavior under the studied conditions.

3.6. Morphology changes in the adsorption process

SEM images, EDS spectra, and elemental mapping are included in Fig. 5(a–d), which shows the morphological and elemental analysis of LDC both before and after heavy metal adsorption. The surface of virgin LDC has a rough, porous, and heterogeneously layered morphology, as seen in Fig. 5a. BET analysis confirmed that the LDC adsorbent possesses a specific surface area of 30.2 m² g⁻¹, and FE-SEM imaging revealed a porous morphology with numerous micro- and macropores. Together, these characterizations demonstrate that the structure provides a considerable surface area and abundant active sites, which are advantageous for adsorption. In accordance with the anticipated composition of the material, the related EDS spectrum verifies the presence of native elements such as oxygen (35.7 wt. %), silicon (19.3 wt. %), calcium (34.2 wt. %), and aluminum (2.1 wt. %). These elements are evenly distributed throughout the surface, as seen in the elemental mapping images.

Upon exposure to Pb²⁺ ions, as seen in Fig. 5b, the LDC surface becomes slightly more compact, likely due to the deposition or complexation of Pb species. The EDS analysis reveals the appearance of a small Pb signal, quantified at approximately 0.1 wt. %, indicating significant adsorption. A slight reduction in Ca and Mg levels suggests potential ion exchange mechanisms. Elemental mapping confirms the homogeneous dispersion of Pb throughout the LDC matrix, particularly in regions also rich in Mg, implying multivalent interactions with surface sites.

Fig. 5c displays the morphological and elemental characteristics of LDC after Co²⁺ adsorption. The SEM image reveals minor surface roughening and possible aggregation, while the EDS data confirm Co presence at 0.2 wt. %. There is also a modest decline in native elements such as Mg and Al, again supporting the likelihood of surface exchange or coordination. Co is distributed uniformly across the surface in the mapping images, which suggests effective and stable interaction of Co²⁺ ions with the LDC framework. Likewise, in Fig. 5(d), following Mn²⁺ adsorption, the LDC retains its overall surface structure but shows minor densification and particle boundary smoothing. The EDS spectrum verifies the presence of Mn at 0.6 wt. %, with little change in other major elemental concentrations. The elemental maps display a consistent Mn distribution, reinforcing the reproducibility and effectiveness of adsorption under these conditions.

Fig. 6 displays the FTIR spectra of LDC before adsorption (black line) and after interaction with Pb²⁺ (red), Co²⁺ (blue), and Mn²⁺ (green) ions. The overall spectral profiles are similar, indicating that the main structural framework of LDC remains preserved after adsorption. However, subtle yet significant shifts in specific absorption bands and

Table 3

The outcomes of kinetics models for three different types of heavy metal adsorption in this study. Condition: pH = 5, Time = 0–120 min, V = 30 mL, Mass of adsorbent = 3.34 g L⁻¹, Pb²⁺-Co = 4.15 mg L⁻¹, Co²⁺-Co = 5.79 mg L⁻¹, and Mn²⁺-Co = 5.52 mg L⁻¹. Pb²⁺: STD_{ave} = 0.22, Co²⁺: STD_{ave} = 0.22, and Mn²⁺: STD_{ave} = 0.31, the number of replications = 2.

PFO model	R ²	RMSE	MAE	k ₁ (min ⁻¹)	Theoretical q _e (mg g ⁻¹)
Pb ²⁺	0.69	0.065	0.05	1	1.26
Co ²⁺	0.89	0.14	0.11	0.095	1.55
Mn ²⁺	0.51	0.15	0.11	0.25	1.02
PSO model	R ²			k ₂ (g.mg ⁻¹ .min ⁻¹)	Theoretical q _e (mg g ⁻¹)
Pb ²⁺	0.97	0.019	0.017	1.99	1.28
Co ²⁺	0.90	0.14	0.1	0.076	1.72
Mn ²⁺	0.76	0.1	0.08	0.49	1.07

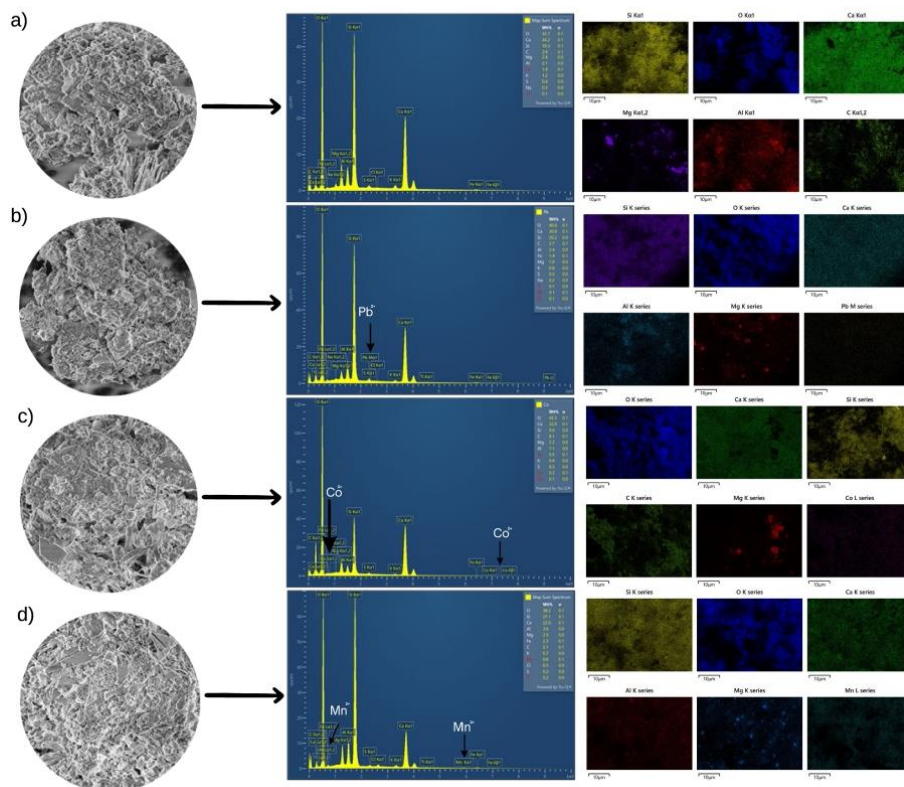


Fig. 5. The results of EDS, elemental mapping and SEM of LDC within interaction of heavy metals based on (a) LDC before adsorption, (b) Pb^{2+} onto LDC, (c) Co^{2+} onto LDC, and (d) Mn^{2+} onto LDC. Conditions: Mass of adsorbent = 3.34 g L^{-1} , $V = 30 \text{ mL}$, $pH = 5$, Time = 30 min, $Pb^{2+}\text{-}C_0 = 4.15 \text{ mg L}^{-1}$, $Co^{2+}\text{-}C_0 = 5.79 \text{ mg L}^{-1}$, and $Mn^{2+}\text{-}C_0 = 5.52 \text{ mg L}^{-1}$.

intensity variations provide evidence of interactions between the functional groups of LDC and the heavy metal ions.

The highlighted region between 900 and 500 cm^{-1} magnified in the inset shows three key peaks at approximately 780 , 680 , and 600 cm^{-1} . These bands correspond to bending vibrations and lattice deformations associated with metal-oxygen (M-O) bonds, such as Si-O-M and Al-O-M linkages, where M represents metal ions (Pb^{2+} , Co^{2+} , or Mn^{2+}). After adsorption, the intensity of these bands increases, and their shapes become slightly sharper, indicating the formation of new metal-associated bonds at the surface (Pasiczna-Patkowska et al., 2025). The band near 680 cm^{-1} is due to overlapping symmetric stretching of Si-O-Mg and Si-O-Al(VI), while the $\sim 600 \text{ cm}^{-1}$ peak indicates M-O-M bond formation. Slight shifts in these peaks after heavy metal adsorption onto LDC confirm surface interactions, evidencing successful adsorption through changes in both the 600 and 680 cm^{-1} regions (Pasiczna-Patkowska et al., 2025; Yan et al., 2012). The peak at 780 cm^{-1} , corresponding to Si-C stretching vibrations (Gualandris et al., 1991), also exhibited noticeable changes after heavy metal (specifically Pb^{2+} and Co^{2+}) adsorption onto LDC, further confirming surface interaction and structural modifications during the adsorption process.

In the higher wavenumber region ($3000\text{--}3700 \text{ cm}^{-1}$), broad bands associated with -OH stretching vibrations remain mostly unchanged, suggesting that the adsorption did not significantly alter the hydration shell or hydroxyl content of the matrix (Su et al., 2009). The sharp peak near 1000 cm^{-1} , typically attributed to Si-O-Si asymmetric stretching

(PI-TSi-GPMS50 structure) (Wahab et al., 2021), also remains consistent across samples, further confirming the structural stability of the LDC framework during metal adsorption.

As illustrated in Fig. 7, the XRD patterns of LDC before to and during heavy metal adsorption showed clear differences throughout the indicated crystallographic planes, indicating various methods of interaction for Mn^{2+} , Co^{2+} , and Pb^{2+} . In Fig. 7a, pristine LDC shows clear peaks at (003), (006), (012), (015), (018), and (113). After adsorption, notable changes occur: Pb^{2+} shifts and intensifies the (006) reflection, indicating interlayer expansion and stronger binding; Mn^{2+} and Co^{2+} shift peaks positively, causing lattice contraction. Peak (003) shows slight d-spacing changes, while (012), (015), and (018) display reduced intensity. Overall, adsorption modifies both peak positions and crystallinity. The XRD results of LDC in Fig. 7a show that heavy metal adsorption causes peak shifts and intensity changes, most notably at the $(006 \approx 2\theta = 26\text{--}27^\circ)$ reflection. A similar trend was reported by Ritonga et al. (2025), where the main red sand peak at $\approx 26^\circ$ decreased after heavy metal adsorption, with a concurrent increase near $\approx 22^\circ$. Both studies confirm that heavy metal incorporation modifies reflection intensities and evidences lattice reorganization. Fig. 7b presents the peak shifts ($\Delta 2\theta$) observed after adsorption of Mn^{2+} , Co^{2+} , and Pb^{2+} on LDC. According to Bragg's law ($n\lambda = 2d \sin \theta$) (Bhattacharjee, 2025), such shifts directly correspond to changes in interplanar spacing (d) (Khan et al., 2020). Fig. 7b shows that Mn^{2+} causes the largest positive peak shift, indicating lattice contraction. Co^{2+} also shifts peaks positively but less

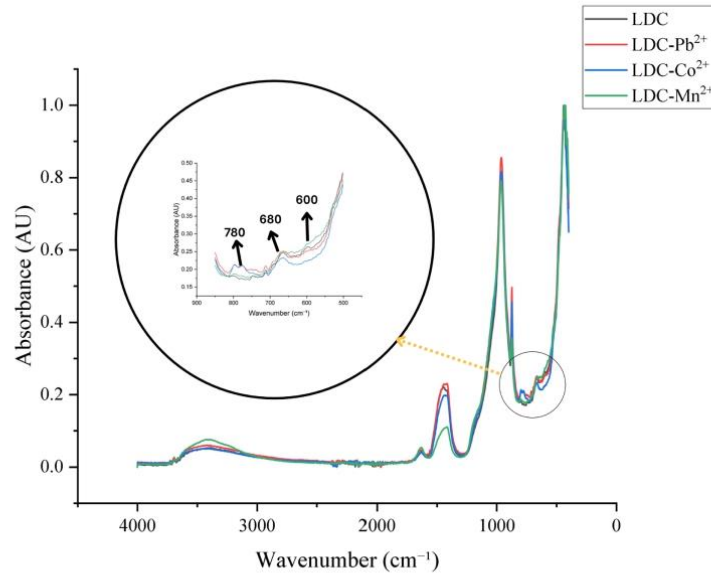


Fig. 6. The FTIR results of LDC in interaction with heavy metals based on LDC before and after adsorption of Pb^{2+} , Co^{2+} and Mn^{2+} onto the surface. Conditions: Mass of adsorbent = 3.34 g L^{-1} , $V = 30 \text{ mL}$, $\text{pH} = 5$, Time = 30 min, $\text{Pb}^{2+}\text{-C}_0 = 4.15 \text{ mg L}^{-1}$, $\text{Co}^{2+}\text{-C}_0 = 5.79 \text{ mg L}^{-1}$, and $\text{Mn}^{2+}\text{-C}_0 = 5.52 \text{ mg L}^{-1}$.

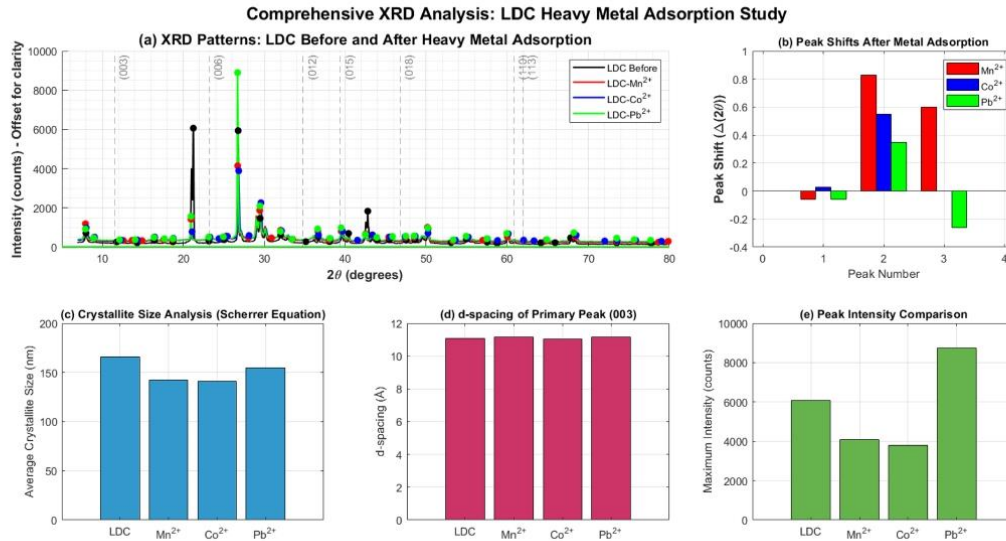


Fig. 7. The outcomes of (a) XRD patterns of LDC before/after Mn^{2+} , Co^{2+} , Pb^{2+} adsorption; (b) peak shifts; (c) crystallite size via Scherrer eq.; (d) d-spacing of (003); and (e) peak intensity comparison. Conditions: Mass of adsorbent = 3.34 g L^{-1} , $V = 30 \text{ mL}$, $\text{pH} = 5$, Time = 30 min, $\text{Pb}^{2+}\text{-C}_0 = 4.15 \text{ mg L}^{-1}$, $\text{Co}^{2+}\text{-C}_0 = 5.79 \text{ mg L}^{-1}$, and $\text{Mn}^{2+}\text{-C}_0 = 5.52 \text{ mg L}^{-1}$. In panel (b): Peak No. 1 → corresponds to the (003) reflection, Peak No. 2 → corresponds to the (006) reflection, and Peak No. 3 → corresponds to the (012) reflection.

strongly, while Pb^{2+} produces slight negative shifts, which can be explained by its smaller hydrated radius that facilitates deeper incorporation into the LDC lattice, thereby expanding the interlayer spacing. These variations confirm that each heavy metal interacts differently with the LDC lattice, altering interplanar spacing and local crystallinity.

Fig. 7c shows the average crystallite size of LDC before and after heavy metal adsorption, determined using the Scherrer equation (Burton et al., 2009). The pristine LDC exhibits the largest crystallite size, whereas the incorporation of Mn^{2+} , Co^{2+} , and Pb^{2+} reduces it, as evidenced by peak broadening that reflects apparent crystallite size

decreases. Microstrain may also contribute to this effect, although it was not deconvoluted during the adsorption process. Mn^{2+} leads to the strongest reduction, indicating higher disruption of crystallite domains. Conceptually, the calculation is based on peak broadening: the full width at half maximum (FWHM) (Burton et al., 2009) of XRD peaks is inversely related to crystallite size, meaning broader peaks correspond to smaller crystallites. Fig. 7d presents the d-spacing values of the primary (003) reflection for pristine and metal-loaded LDC, calculated from Bragg's law. Here, the 2 θ positions extracted from peak analysis were converted into interplanar spacings (d), where smaller $\Delta 2\theta$ yields larger d-values. The results show that adsorption of Pb^{2+} slightly increases d-spacing, consistent with local interlayer expansion, while Mn^{2+} and Co^{2+} cause minor reductions, reflecting lattice contraction. Fig. 7e compares the maximum peak intensities, determined by selecting the highest counts in each diffractogram. The pristine LDC exhibits intermediate intensity, which decreases significantly after Mn^{2+} and Co^{2+} adsorption, suggesting increased disorder (Babu and Seehra, 1996), while Pb^{2+} produces enhanced intensity, implying stabilization or preferred orientation of certain crystallographic planes.

3.7. Competition effects

Fig. 8 presents the results of a competitive adsorption study designed to evaluate the performance of LDC in removing Pb^{2+} , Co^{2+} , and Mn^{2+} ions under varying initial concentrations. The study examines three distinct scenarios low, middle, and high concentrations and compares both the adsorption capacity (Fig. 8a) and removal efficiency (Fig. 8b) for each ion. The aim is to understand the influence of co-existing metal ions on the selective removal of Pb^{2+} , which is considered the primary contaminant in this background.

In Fig. 8a, the adsorption capacity (q_e , mg g^{-1}) for Pb^{2+} is consistently higher than for Co^{2+} and Mn^{2+} across all concentration scenarios. At low concentrations, Pb^{2+} exhibits a q_e of approximately 6.0 mg g^{-1} , while Co^{2+} and Mn^{2+} show much lower uptake values of around 1.5 and 1.2 mg g^{-1} , respectively. As the concentration increases, this trend becomes more pronounced: at middle concentrations, Pb^{2+} reaches roughly 7 mg g^{-1} , and at high concentrations, the capacity increases significantly to nearly 14.5 mg g^{-1} , although with greater variability as indicated by the error bars. In contrast, Co^{2+} and Mn^{2+} exhibit smaller incremental increases, with Co^{2+} achieving a maximum of about 6.7 mg g^{-1} and Mn^{2+} remaining under 3.34 mg g^{-1} . These results indicate a preferential affinity of LDC for Pb^{2+} , even in the presence of competing ions and at

elevated concentrations. Fig. 8b complements these findings by illustrating the removal efficiency (%) for each ion across the same three scenarios. In the low-concentration condition, Pb^{2+} is removed with the highest efficiency approaching 95 % while Co^{2+} and Mn^{2+} achieve about 45 % and 38 % removal, respectively. This selectivity diminishes at higher concentrations: at middle levels, removal efficiencies converge to around 17–25 % for all ions, suggesting a saturation of active sites and intensification of competitive interactions. At high concentrations, Pb^{2+} removal drops to nearly 25 %, while Co^{2+} and Mn^{2+} follow closely at 23 % and 16 %, respectively. Despite the decreasing trend in percentage removal, Pb^{2+} still maintains a slightly higher removal rate, reinforcing its dominant adsorption behavior on the LDC surface. Together, these results demonstrate that Pb^{2+} consistently outcompetes Co^{2+} and Mn^{2+} in terms of both adsorption capacity and removal efficiency under all tested conditions. The preferential uptake of Pb^{2+} is attributed to its higher initial concentration, potentially stronger binding affinity, and favorable physicochemical interactions with LDC functional groups. Notably, the differences in adsorption behavior become more pronounced at lower concentrations, where active sites are more available and selective adsorption dominates. As concentrations increase, competition intensifies, leading to a relative decline in selectivity and efficiency.

In other studies, it has been demonstrated that the adsorption rate and binding strength of heavy metal cations are significantly influenced by factors such as ionic radius and, more importantly, their hydrated radius. These parameters play a critical role in determining the selectivity of adsorption, in conjunction with the specific nature and characteristics of the adsorbent surface, particularly the chemical and structural features of the adsorption centers (Martemianov et al., 2017). For instance, Inyang et al. justified the preferential retention of Pb^{2+} over Cd^{2+} on both studied clay types by highlighting the smaller hydrated radius of Pb^{2+} . This smaller radius enables a more intimate and stable interaction with the clay surface, offering a better spatial fit compared to the larger cadmium ions, thereby enhancing Pb^{2+} selectivity (Inyang et al., 2016).

3.8. Adsorbent regeneration prediction model (ARPM)

The adsorption of heavy metals onto LDC was tested for five replicate sorption-desorption cycles, using $1 \text{ mol L}^{-1} \text{ HNO}_3$ as the desorbing solution. The results of the experiments (Fig. 9a and b) show that the adsorption percentage of heavy metals by LDC linearly decreased with

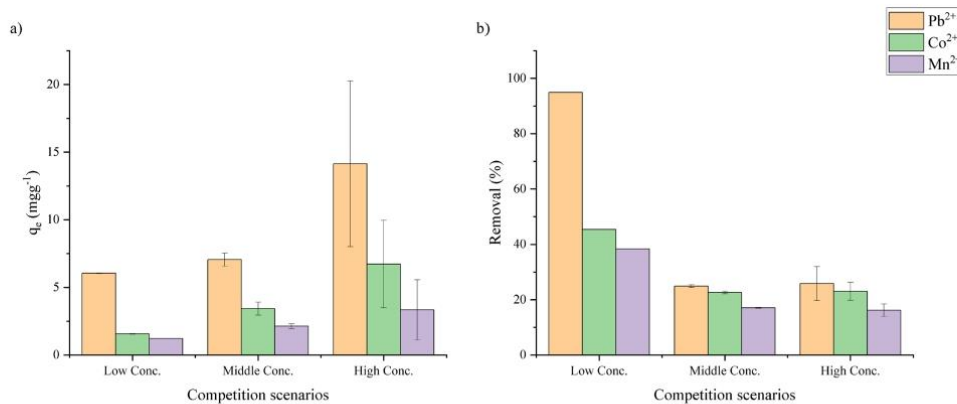


Fig. 8. The results of competitive adsorption of Pb^{2+} , Co^{2+} , and Mn^{2+} onto in different scenarios based on (a) adsorption capacity and (b) removal (%). Conditions: Mass of adsorbent = 3.34 g L^{-1} , $V = 30 \text{ mL}$, $\text{pH} = 5$, Time = 30 min. low concentrations: $\text{Pb}^{2+}\text{-C}_0 = 21.24 \text{ mg L}^{-1}$, $\text{Co}^{2+}\text{-C}_0 = 11.42 \text{ mg L}^{-1}$, and $\text{Mn}^{2+}\text{-C}_0 = 10.54 \text{ mg L}^{-1}$, middle concentrations: $\text{Pb}^{2+}\text{-C}_0 = 94.38 \text{ mg L}^{-1}$, $\text{Co}^{2+}\text{-C}_0 = 50.56 \text{ mg L}^{-1}$, and $\text{Mn}^{2+}\text{-C}_0 = 41.64 \text{ mg L}^{-1}$, and high concentrations: $\text{Pb}^{2+}\text{-C}_0 = 182.15 \text{ mg L}^{-1}$, $\text{Co}^{2+}\text{-C}_0 = 97.22 \text{ mg L}^{-1}$, and $\text{Mn}^{2+}\text{-C}_0 = 68.67 \text{ mg L}^{-1}$, the number of replications = 2.

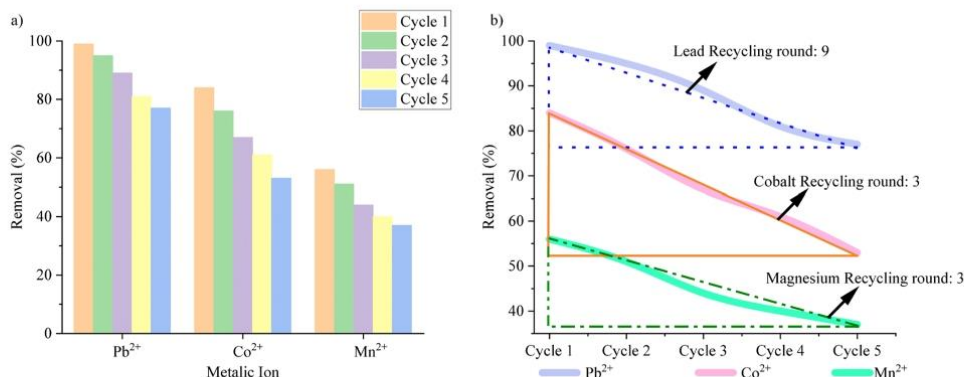


Fig. 9. The test of reusability of the LDC for the removal of heavy metals based on (a) Removal Percentage vs. cycle as bar charts and (b) Thales-based theorem. Conditions: pH, contact time, and the mass: optimal amounts and Desorbing solution: 1.0 mol L⁻¹ HNO₃.

number of sorption cycles. The largest decrease in removal efficiency was observed for Co²⁺ (~7.7% loss per cycle), and lower for Pb²⁺ (5.8 %) and Mn²⁺ (4.9 %). Therefore, LDC can be effectively used as an adsorbent for the treatment of heavy metal ions in repeated cycles.

Zhou and Haynes (2011) explored the mechanisms and conditions affecting desorption by acids. They found that low pH levels facilitate the release of metal cations, as acidic conditions promote the dissolution of Fe and alkoxy/silicate adsorption surfaces, leading to the release of adsorbed metals and surface deposits. Additionally, the strong competition between H⁺ ions and metal cations for adsorption sites causes the displacement of cations in acid solutions. Moreover, acids react with residual alkalinity, which reduces adsorption capacity (Zhou and Haynes, 2011). However, the exact mechanism of repulsion was not explained in their study. Therefore, the reduction of adsorption capacity after a specific cycle can be related to reactions between acidic and basic environments in the present research. In a related study, Iqbal et al. (2002) investigated the removal of heavy metals from PFP using 0.1 M HCl over three cycles. They found that the removal efficiency for most of the heavy metals studied was over 90 %.

The current study employed the ARPM computational method to calculate the cycle of LDC regeneration for each metal ion, as presented in Table 4. The Cs values are standard allowable concentrations of heavy metal ions in river water as per World Health Organization (WHO) protocol (Rozana et al., 2020; Waghmare et al., 2015). Additionally, the initial concentration of heavy metals (C₀) in the effluent of a wastewater treatment plant located in Toos industrial park (Mashhad, Iran) was demonstrated in Table 4, based on field surveying.

Based on ARPM results presented in Table 4, it is recommended to regenerate LDC for Co²⁺ and Mn²⁺ after a minimum of three cycles of adsorption to ensure resilience in sequential reactions. The maximum regeneration cycle with nine sequential reactions is associated with Pb²⁺. Therefore, it can be concluded that ARPM is suitable for predicting the cycle of regeneration for each adsorbent and can provide satisfactory

results. The validation of the extended ARPM model is illustrated using experimental data for Co²⁺ and Mn²⁺ in Fig. S5. For Co²⁺ with an initial concentration of 0.15 mg L⁻¹, the concentration decreased to 0.07 mg L⁻¹ after five adsorption-regeneration cycles. By the third cycle, the concentration reached 0.0495 mg L⁻¹, nearly equal to the standard limit of 0.05 mg L⁻¹. A similar trend was observed for Mn²⁺ (C₀ = 0.9 mg L⁻¹), which declined to 0.504 mg L⁻¹ after three cycles, aligning with the threshold of 0.5 mg L⁻¹. Beyond these points, further cycles would result in effluent concentrations exceeding acceptable limits. These results validate the ARPM framework, demonstrating its ability to accurately predict regeneration limits based on key parameters: initial ion concentration, water quality standards, the linearity of regeneration decline, and cycle count.

It is worth noting that the calculations and mathematical models outlined in this section of research offer a valuable tool to determine the regeneration cycle of LDC when faced with unknown water or wastewater samples. By simply inputting the initial concentration values of heavy metals found within the samples, one can precisely calibrate the minimal time for the regeneration. This functionality holds immense significance not only for LDC but can also be extrapolated to various adsorbent types and their interactions with different pollutants. This capability becomes particularly important in waste materials applications, where real-world performance data can greatly enhance water and wastewater treatment processes. Understanding how these materials perform in practical settings allows for the maintenance of peak operational efficiency. Given the cost considerations associated with replacing materials, especially within the operation section of water treatment systems, preserving the longevity of adsorbents becomes paramount. By leveraging the insights garnered from these models, operators can effectively manage resources while upholding high treatment standards.

The regeneration of LDC adsorbents using acids such as HNO₃ involves three primary mechanisms, as illustrated in Fig. S6. The first mechanism, proton exchange, occurs when HNO₃ introduces H⁺ ions that effectively replace adsorbed metal ions on the surface of the adsorbent. This process is facilitated by the protonation of surface functional groups in the presence of water molecules, leading to the displacement of metal ions (M⁺) from the active sites (Renu and Sithole, 2024). The second mechanism, pH-induced desorption, involves structural changes caused by the acidic environment (Renu and Sithole, 2024). A lowered pH alters the surface charge and disrupts electrostatic or coordination bonds between the metal ions and oxides like SiO₂, Al₂O₃, and CaO (refer to XRF characterization of LDC), resulting in the release of metals. Lastly, in the solubilization step, desorbed metals are dissolved into the regenerant solution, restoring the active sites of the

Table 4

The initial concentration of heavy metal, the allowable standard concentration of heavy metals and the cycle of adsorbent regeneration, C₀: Initial concentration of heavy metal ions and Cs: standard allowable concentrations of heavy metal ions in river water as per WHO protocol (Rozana et al., 2020; Waghmare et al., 2015).

Ions	Pb ²⁺	Co ²⁺	Mn ²⁺
C ₀ (mg L ⁻¹)	0.025	0.15	0.9
Cs (mg L ⁻¹)	0.01	0.05	0.5
Regeneration Cycle	9	3	3

adsorbent for reclamation (Renu and Sithole, 2024). Together, these mechanisms ensure efficient regeneration and reutilization of the LDC material in successive treatment cycles.

Regeneration of adsorption using acids provides high efficiency in desorbing heavy metals, as acidic agents such as HCl, HNO₃, and H₂SO₄ effectively dissolve metal-adsorbent (e.g., biochar) complexes and reduce pore blockage from insoluble precipitates, thereby restoring adsorption capacity and occasionally enlarging the surface area to expose new active sites. This makes acid treatment generally superior to alkaline agents in the initial cycles, delivering strong desorption performance. However, repeated acid use presents notable drawbacks, as strong acids, particularly HNO₃, degrade the adsorbent structure by dissolving minerals and damaging functional groups, which progressively reduces adsorption efficiency. Moreover, acid regeneration produces secondary effluents rich in metals and acidity, posing risks of secondary pollution and necessitating additional treatment. In industrial applications, this drawback becomes especially critical. Thus, while acids enhance regeneration efficiency, they compromise long-term stability and sustainability, which should be considered a limitation of the present investigation (Alsawy et al., 2022).

Fig. S7(a-d) provide a comprehensive view of regeneration dynamics for Pb²⁺, Co²⁺, and Mn²⁺ in water treatment operations. Fig. S7(a) illustrates the efficiency decline across cycles, enabling managers to plan regeneration before performance drops below acceptable levels. Fig. S7(b) compares influent concentrations with WHO standards, where Mn²⁺ emerges as a priority pollutant requiring strict control. Fig. S7(c) highlights average relative decline rates, stressing the need for material selection and scheduling strategies to extend adsorbent lifetime. Finally, Fig. S7(d) presents a heatmap of removal performance, where critical low-performance zones are easily identified, guiding proactive maintenance. Collectively, these results equip plant managers with actionable insights, linking laboratory findings to operational planning, regulatory compliance, and cost-effective regeneration management.

3.9. Sustainable materials management model (SMM)

Various adsorbents are used for heavy metal decontamination, with GO/Carbon commonly employed. Therefore, this research compares the applied adsorbent to the most commonly used materials, such as GO/Carbon. The outcomes of SMM analysis for both GO/Carbon and LDC adsorbents are demonstrated in Fig. 10. The quantitative data for LDC and GO/Carbon materials regarding their environmental impacts are summarized in Table S4, while the results of the indicators in both supply chain and operational categories are presented in Table S5. As per Fig. 10a, it can be understood that in the impact potential category,

some indexes of LDC, including Acid Rain (ACID), Freshwater Aquatic Ecotoxicity (ETOX), Human Health – Respiratory Effects (HRSP), Human Health – Toxicity (HTOX), Ozone Depletion (OZON), and Smog Formation (SMOG) were more appropriate than GO/Carbon adsorbent. While in the category, the maximum difference was related to ETOX and HTOX with 64 % and 51 %, respectively. Therefore, it can be concluded that from toxicity aspects, the LDC is safer in comparison of GO/Carbon adsorbents. Likewise, the analysis illustrated that the Eutrophication (EUTR) index of GO/Carbon is much lower than LDC adsorbent (Kim and Chae, 2016).

Also, in the category of Resource Use, water (WATR) and energy (ENRG) consumptions of LDC performed better than GO/Carbon with 12 % and 44 % difference, correspondingly. However, the computations demonstrate that the Minerals and Metals Use (MNRL) and Land Use (LAND) values of GO/Carbon were enormously lower than LDC one. However, there is a sensitive point about the comparison of both the materials as adsorbents. The GO/Carbon should be synthesized from raw materials, while the LDC which is used in this study is waste material of CDW category. It can be understood that with the assumption, both indexes of MNRL and LAND of LDC could not be worse than GO/Carbon because no specific area and minerals are applied for fabricating the wastes.

From the Commercial Resource Conservation and Recovery Act-RCRA Hazardous Waste (CRHW) aspect, the LDC was 26 % better than GO/Carbon adsorbent as per the computations. However, for both Commercial Construction and Demolition Debris (CCDD) and Commercial Municipal Solid Waste (CMSW), the model showed that the GO/Carbon is better than LDC. In the simulation, the LDC is defined as a concrete product, but in reality, it is a waste of concrete product. Therefore, considering the waste management practices of LDC, the overall condition of waste generated (CRHW, CCDD, and CMSW) in the LDC category would be better than that of GO/Carbon.

Fig. 10b and c show the environmental impacts of different SMM indexes in both LDC and GO/Carbon cases as per supply chain and operational categories. Supply Chain refers to the full set of processes required to provide a good or service, whereas Operations handles problems that can occur from the releases of production, the use of resources, and the creation of waste at the facilities (US EPA, 2020).

The analysis of the supply chain (Fig. 10b) demonstrates that just in water consumption and CMSW, concrete products have better condition in comparison to GO/Carbon. The LDC waste is not initial material, but it is repurposed CDW, which is applied again for water purification. In the following, these values of environmental indicators and their effects can be adjusted with management coefficients and rough estimates. Fig. 10c demonstrated that in most environmental impact factors, the

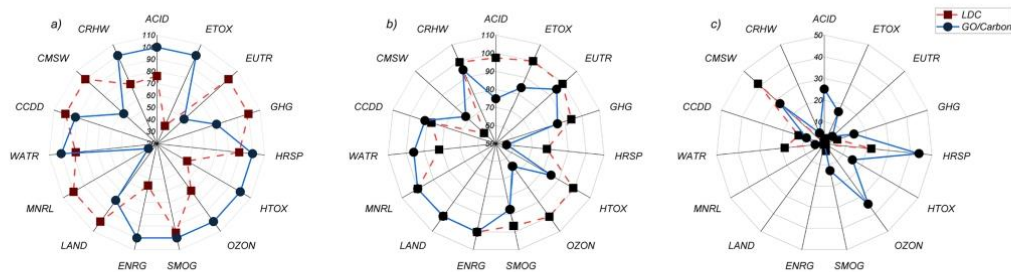


Fig. 10. The outcomes of SMM analysis based on (a) environmental impacts, (b) supply chain contribution (%), and (c) operational contribution (%) in different categorizations. Description: Acid Rain (ACID) (kg SO₂-eq); Freshwater Aquatic Ecotoxicity (ETOX) (CTUe); Eutrophication (EUTR) (kg N-eq); Greenhouse Gases (GHG) (kg CO₂-eq); Human Health – Cancer (HCAN) (CTUh); Human Health – Noncancer (HNCN) (CTUh); Human Health – Respiratory Effects (HRSP) (kg PM_{2.5}-eq); Human Health – Toxicity (HTOX) (CTUh); Ozone Depletion (OZON) (kg CFC-11-eq); Smog Formation (SMOG) (kg O₃-eq); Energy Use (ENRG) (MJ); Land Use (LAND) (m²-yr); Minerals and Metals Use (MNRL) (kg); Nonrenewable Energy Use (NNRG) (MJ); Renewable Energy Use (RNRG) (MJ); Water Use (WATR) (m³); Hazardous Air Pollutants (HAPS) (kg); Metals (METL) (kg); Pesticides (PEST) (kg); Commercial Construction and Demolition Debris (CCDD) (kg); Commercial Municipal Solid Waste (CMSW) (kg); Commercial RCRA Hazardous Waste (CRHW) (kg); Jobs Supported (JOBS); Value Added (VADD) (\$).

LDC wastes are more appropriate than GO/Carbon. Just about LAND, WATR, CCDD, and CMSW GO/Carbon were more suitable than the concrete products. However, considering the fundamental waste of the LDC, it can be concluded that in all the indexes, the condition of LDC is better than GO/Carbon.

Scaling up LDC production from AAC waste may involve challenges such as ensuring material consistency and managing energy inputs. For managing spent LDCs loaded with heavy metals, incorporation into cement-based solidification/stabilisation (S/S) systems is an effective strategy. As described by [Chen et al. \(2009\)](#), heavy metals interact with cement hydration products via sorption, precipitation, and chemical incorporation. For example, calcium-silicate-hydrate (C-S-H) gels serve as key binding phases. Heavy metals may also form double hydroxides with calcium, promoting further hydration. These mechanisms significantly reduce leachability, making cement-based S/S a viable method for stabilizing metal-loaded cement-based materials like LDC and preventing environmental contamination ([Chen et al., 2009](#)). [Masood et al. \(2001\)](#) propose recycling demolished concrete waste by partially replacing cement and fine aggregates in new mixes. Their results show that up to 20 % replacement maintains 75–77 % of conventional concrete strength. This strategy reduces the use of natural aggregates and provides environmental and economic benefits, supporting waste concrete as a viable construction material.

Scaling up adsorbent pervious concrete for commercial applications presents significant technical challenges that must be carefully managed. The research of [Teymouri et al. \(2025\)](#) indicates that optimal mix designs typically require aggregate contents of 1400 kg/m³ using 4.75–9.5 mm sized particles, cement contents of 280–330 kg/m³, and water-to-cement ratios between 0.25–0.45 to achieve the necessary balance between structural integrity and filtration performance. The porosity must be maintained within 15–25 % to ensure adequate permeability (0.14–1.22 cm/s) while preserving compressive strength in the 3–28 MPa range, though these properties follow an inverse relationship with correlation coefficients up to $R^2 = 0.92$. Contamination removal efficiency can reach to acceptable values for heavy metals and 90 % for Chemical Oxygen Demand/ Biochemical Oxygen Demand (COD/BOD) when fine adsorbents like zeolite or LECA are incorporated at 5–15 % replacement levels. However, long-term performance data reveals that infiltration rates decline approximately 10 % annually for the first four years due to clogging, necessitating maintenance protocols. The scale-up process requires precise quality control systems to maintain consistent aggregate gradation, proper mixing sequences to ensure uniform adsorbent distribution, and field curing procedures that preserve the interconnected pore structure essential for both structural performance and water treatment effectiveness.

[Table 5](#) presents a comparative evaluation of various cement-based adsorbents for heavy metal removal, focusing on adsorption capacity, optimal conditions, and kinetics. Among the materials, vermiculite concrete modified with ferric oxyhydroxide exhibited the highest Pb²⁺ adsorption capacity at 197.6 mg g⁻¹, reflecting its strong binding potential ([Martemianov et al., 2017](#)). In contrast, crushed concrete

fractions (CCF) demonstrated a lower capacity for Pb²⁺ at 37.9 mg g⁻¹ ([Coleman et al., 2005](#)), though still notable, while LDC achieved 43.1 mg g⁻¹ for Pb²⁺, 15.2 mg g⁻¹ for Co²⁺, and 23.5 mg g⁻¹ for Mn²⁺, confirming its broad applicability. In terms of kinetics, LDC outperformed all others with a high rate constant of 1.99 g mg⁻¹ min⁻¹ for Pb²⁺, indicating very rapid adsorption, compared to moderate values for Concrete/maghemite nano material (CMnano) (0.524 g mg⁻¹ h⁻¹ = 0.008 g mg⁻¹ min⁻¹) and vermiculite concrete (0.299 g mg⁻¹ min⁻¹). Operational pH varied across materials, with most functioning at pH 5, except CCF, which performed optimally at pH 11.9 ([Coleman et al., 2005](#)) due to its alkaline nature. Additionally, LDC achieved complete adsorption within 20–30 min (Co²⁺ and Pb²⁺: 20 min, Mn²⁺: 30 min), making it highly suitable for time-sensitive treatment, while other materials required prolonged contact times (e.g., 48 h for CCF, 120 min for vermiculite concrete, 24 h for waste cement powder, and 40 h for CMnano). Also, it is worth noting that all adsorbents followed second-order kinetics. Overall, LDC from the present study offers a promising balance of efficiency, speed, and versatility across different heavy metal contaminants.

CDWs have been widely applied for heavy metal adsorption in various applications, most of which involve soil-based components such as CaO and SiO₂ ([Ranaweera et al., 2023](#)). In contrast, LDC is designed to achieve lighter weight with structural engineering benefits ([Chen et al., 2019](#)). To meet this objective, materials such as Al₂O₃ are incorporated into the LDC mixture, which increases porosity. Enhanced porosity facilitates improved mass transfer mechanisms for heavy metals and other contaminants, thereby strengthening the adsorption performance ([Gong et al., 2023](#)). Additionally, the addition of Al₂O₃ not only alters the physical structure but also influences the chemical specification of LDC, resulting in additional effects on the adsorption process, particularly for heavy metals ([Mola-Abasi et al., 2018](#)). Thus, the preparation of LDC originally intended for structural engineering purposes such as light-weight construction and improved seismic performance also provides both physical and chemical advantages on the adsorbent surface, making it highly effective for heavy metal decontamination.

Finally, the limitation of the present study are:

- The ARPM was validated only under controlled laboratory conditions; external or cross-system validation is still required in future studies.
- Acid-based regeneration suggests the Thales model applies mainly under physical sorption with linear reduction, limiting its applicability in non-linear conditions.
- Experiments were conducted in duplicate ($n = 2$), which may limit reproducibility and should be validated with higher replication in future work.

4. Conclusion

According to the principles of integrated solid waste management, the utilization of waste materials is an important issue. In this study, AAC waste, which is one of the primary construction wastes in Iran,

Table 5
The summary of CDW applications as adsorbent for decontamination of heavy metals in different studies.

Adsorbent	Contamination	Adsorption capacity (mg.g ⁻¹)	Optimal contact time (min)	Operational pH	Kinetics order	Kinetics Coefficient (g.mg ⁻¹ .min ⁻¹)	Ref.
Waste cement powder	As ³⁺	28.3	1440	10–11	—	—	(Sasaki et al., 2014)
Concrete/maghemite nano material (CMnano)	As ³⁺	11.12	2400	5	PSO	0.008	(Hernández-Flores et al., 2018)
Vermiculite concrete modified with Ferric oxyhydroxide	Pb ²⁺	197.6	120	5	PSO	0.299	(Martemianov et al., 2017)
Crushed Concrete Fractions (CCF)	Pb ²⁺	37.9	2880	11.9	—	—	(Coleman et al., 2005)
LDC	Pb ²⁺	43.1	20	5	PSO	1.99	The present study
	Co ²⁺	15.2	20	5	PSO	0.076	
	Mn ²⁺	23.5	30	5	PSO	0.49	

India, China, and other developing countries, was modified to create an efficient adsorbent for the removal of heavy metals and converted into LDC. This approach aligns with some environmental platforms, such as Sustainable Development Goals (SDGs), which promote the use of waste materials as adsorbents. The analysis demonstrated that the optimal pH of 5 and minimal mass of adsorbent 6.66 gL^{-1} for adsorption of Pb^{2+} , Co^{2+} , and Mn^{2+} onto LDC were equal. Also, the minimum contact time for reasonable equilibrium adsorption of Pb^{2+} , Co^{2+} and Mn^{2+} are 20 min and 30 min, respectively. The isothermal modeling demonstrated that Pb^{2+} , Co^{2+} , and Mn^{2+} adsorption onto LDC wastes occurs physically as multilayer adsorption on a heterogeneous surface. Moreover, the kinetic analysis confirmed that the adsorption of Pb^{2+} onto LDC occurs faster than that of Mn^{2+} and Co^{2+} , and the reactions follow a PSO mechanism. Moreover, the competitive adsorption tests revealed that LDC exhibits a higher selectivity for Pb^{2+} compared to the other ions. According to the ARPM, lead has the highest regeneration threshold of nine cycles among all the heavy metal ions under study, while cobalt and manganese have the lowest and is not advised for more than three regeneration cycles. Furthermore, EPA's SMM model indicates that LDC waste is more sustainable than other adsorbents like GO and carbon-based materials.

In future studies, evaluating the efficacy of waste materials in heavy metal decontamination procedures may benefit from the use of machine learning algorithms. Also, in this study, acid-based regeneration indicates that the Thales-based model applies mainly where physical sorption dominates, leading to expected linear reduction trends. However, the model's lack of applicability in non-linear conditions could be considered a limitation, and this hypothesis requires further validation in future studies. Moreover, next investigations could validate heavy metal removal by LDC in real wastewater, assess risks of leaching from spent LDC, and conduct full Life Cycle Assessment comparing its sustainability with other adsorbent materials.

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CRediT authorship contribution statement

Mohammad Gheibi: Writing – original draft, Visualization, Software, Project administration, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Seyyed Roohollah Masoomi:** Writing – original draft, Software, Methodology, Investigation, Conceptualization. **Mohammad Eftekhari:** Writing – original draft, Validation, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Mehran Akrami:** Methodology, Conceptualization. **Martin Palusák:** Validation, Methodology, Conceptualization. **Daniele Silvestri:** Writing – review & editing, Supervision, Project administration, Conceptualization. **Miroslav Černík:** Writing – review & editing, Supervision, Project administration, Funding acquisition. **Stanisław Wacławek:** Writing – review & editing, Supervision, Project administration, Methodology.

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.

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Supplementary materials

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Data availability

Data will be made available on request.

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4.2. Automation system design for heavy metal adsorption onto CDWs

4.2.1. Sustainable and Intelligent Heavy Metal Adsorption onto LDC

This study presents a smart, Industry-4.0-oriented framework for heavy-metal removal from water using LDC waste as a sustainable adsorbent. Experimental adsorption of Pb^{2+} , Co^{2+} , and Mn^{2+} was combined with Response Surface Methodology (RSM) and 20 machine-learning algorithms to model, optimize, and control removal efficiency. Key operational variables included pH, metal type, initial concentration, adsorbent mass, and contact time. Results identified pH and metal type as dominant factors, with optimal performance consistently achieved at $\text{pH} \approx 5$ and adsorbent dosages of $8.67\text{--}10 \text{ g L}^{-1}$. Among all models, Tree Random Forest (TRF), lazy IBK, and multilayer perceptron showed the highest predictive accuracy (correlation coefficient > 0.92), with TRF reaching ~ 0.99 under optimized training conditions. A major contribution is the integration of the optimized ML model into a Knowledge Flow-based decision support system, enabling real-time prediction, classification, and alarm-based process control for adsorption systems. This transforms adsorption from a static, laboratory-scale technique into a digitally enabled, adaptive treatment process suitable for smart water infrastructure. Main targets of the study are: (i) sustainable reuse of construction waste within a circular-economy framework, (ii) intelligent optimization and automation of heavy-metal adsorption, and (iii) real-time decision support for water treatment operations. The work directly contributes to **Research Objective 4**, namely the development of AI-driven, data-integrated control systems for resilient, resource-efficient environmental technologies.



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Smart control of heavy metal adsorption onto LDC wastes for Industry 4.0 applications

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ABSTRACT

Due to the complexity and the need for automation in adsorption systems, this study develops a decision-making model using 20 Machine Learning Algorithms (MLAs), Response Surface Methodology (RSM), and lab tests. Inputs include pH, metal type, concentration, adsorbent mass, and time; output is removal percentage for process control. The best-performing models (Tree Random Forest: TRF, lazy Instance-Based K: IBK, and Function Multilayer Perceptron: FMLP) achieve high accuracy for prediction of Removal Percentage (RP) of heavy metals with >0.92 correlation coefficient and >0.8 recall/precision indicators. As a novelty of this study, a decision-making model for the heavy metal adsorption process onto waste materials is developed for the first time using the Knowledge Flow (KF) platform in WEKA software, incorporating real-time data to improve process control and operational efficiency. The results demonstrated that the cation type and pH, with a P-value < 0.001 , are the most significant factors affecting the RP. To achieve maximum RP, the pH should be set to 5, and the adsorbent amount should be in a range of $8.67\text{--}10\text{ g L}^{-1}$, regardless of the initial concentration and type of ions (Pb^{2+} , Mn^{2+} , and Co^{2+}). Then, evaluating MLAs showed that TRF, with a correlation coefficient exceeding 0.96, performs best for predicting RP, potentially reaching 0.99 at a split percentage around 80%. TRF uses real-time experimental data in water treatment systems to anticipate RP up to 98.75% and to activate alarms for RP below 80% using the KF principle.

1. Introduction

The World Health Organization (WHO) reports that 844 million people lack access to a basic supply of drinking water, and that 230 million people spend more than 30 minutes per day collecting water from improved sources, including piped water, boreholes, protected wells and springs, rainwater, and packaged or delivered water (WHO, Edition, 2011; Joseph et al., 2019; UNICEF, 2017). In developing countries such as Angola, Algeria, and Cameroon, increasing pollution is especially concerning because people lack the resources to treat polluted water or access clean drinking water (López and Toman, 2006). The problem of water quality is not limited to developing countries; even some developed countries, such as Italy, face significant issues due to heavy metals in their water resources (Triassi et al., 2023).

Importantly, the impact of water contamination extends beyond the immediate environment, affecting food sources and other parts of the community farther from the water source. Vegetables and fruits from a suburban industrial area in Bangladesh were found to contain elevated levels of heavy metals, including As, Cd, Pb, Mn, and Fe. Health risks were significant, particularly in relation to cancer, indicating unsuitability for growing certain vegetables (Sultana et al., 2017). As such, the discharge of heavy metals has a negative effect on the food chain. It is considerably more effective to control contamination in water resources than to deal with it later (Arunakumara and Zhang, 2008).

Adsorption (Arora, 2019), coagulation (Charemtanyarak, 1999), electrocoagulation (Bazrafshan et al., 2015), membrane technologies (Qdais and Moussa, 2004) and catalytic treatments (Das and Poater, 2021) are among the various techniques for decontaminating heavy

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metals; each has pros and cons of its own. The adsorption method is ideal due to its low energy use, low cost, and ease of operation (Abas et al., 2013). While adsorption is inexpensive, it is not automated; chemical treatment is also inexpensive but polluting; electric-based treatment is more environmentally friendly but requires more energy and costs; membranes are clean but expensive; and photocatalysis offers notable environmental benefits; however, it is not highly cost-effective. Combining treatment techniques can enhance overall process efficiency and removal performance (Qasem et al., 2021). Therefore, they have been extensively targeted as viable methods for heavy metal decontamination in numerous studies. However, critical factors such as the type of adsorbent, adsorption mechanisms, and the incorporation of intelligent operational strategies remain important considerations and represent existing research gaps. Moreover, when waste materials are selected as adsorbents within an Integrated Waste Management System (IWMS) framework, the complexity of the operational process increases, requiring more advanced optimization and control approaches. The application of waste materials such as Construction and Demolition Waste (CDW) as an adsorbent could be an example of circular economy principles in water treatment processes (Gheibi et al., 2025).

CDW consists of materials produced during construction, renovation, and demolition activities, including concrete, brick, sand, and gravel. The widespread use of concrete is drawing attention to inexpensive calcium-based adsorbents made from concrete waste. In the face of economic and environmental concerns, sustainable CDW reuse is crucial due to growing demand (Pallewatta et al., 2023). In Gheibi et al., CDW-derived adsorbents show complex heavy metal adsorption behavior because their surfaces are highly heterogeneous and porous, containing varied oxides such as CaO, SiO₂, and Al₂O₃. These irregular surface energies and diverse functional sites lead to multilayer and non-uniform adsorption, making the interaction between heavy metals and CDWs inherently complex (Gheibi et al., 2025). Due to this complex CDW behavior, statistical methods such as Response Surface Methodology (RSM) help assess key factors and predict adsorption process effectiveness (Anfar et al., 2020). RSM aims to build regression models for predicting responses, though accuracy may vary. However, these statistical methods are inherently static by design, and for dynamic analysis and real-time operation of adsorption systems, more advanced intelligent models based on artificial intelligence can be highly beneficial. Machine Learning Algorithms (MLAs) enhance prediction, offering smarter, more precise adsorption process control and enabling expert systems for efficient water purification (Han et al., 2025; Yaseen, 2021).

Pardakhti et al. (2017) employed MLAs to lessen the computational burden of molecular simulations for adsorbent screening. They used a range of machine learning algorithms, such as decision trees and support vector machines, to estimate methane uptake in Metal-Organic Frameworks (MOFs). In their study, 92 % of the dataset (130,398 MOFs) is used for training the model, while 8 % is used for testing. Yang et al. (2021) used 4420 data points from 1105 soils to create MLAs for heavy metal adsorption on soil. They discovered that the gradient boosting algorithm is the best decision tree algorithm for forecasting the adsorption of six heavy metals. To predict adsorption abilities and pollutant levels in water, independent and combined models are made. The Shapley additive explanation method helps identify important features. In their study, Shapley Additive Explanations (SHAP) are applied to interpret machine-learning models by quantifying how each input feature contributes to a prediction, enabling transparent assessment of variable importance, model behavior, and decision-making in complex environmental systems. Fanourgakis et al. (2019) proposed new descriptors for MLAs to predict the gas uptake capacities of nanoporous materials. Using void fractions of particles with defined van der Waals radii as descriptors, they assessed the random forest algorithm on a dataset of 4700 experimental MOFs. On the other hand, the results demonstrated that the proposed approach is generalizable beyond MOFs to other nanoporous materials. Zhu et al. (2021) apply MLAs to predict the adsorption of antibiotics on Carbon-based Materials (CBMs).

Comparing random forest, gradient boosting trees, and Artificial Neural Networks (ANN), they found the random forest models most accurate. Relative importance analysis and partial dependence plots highlight key factors affecting adsorption, proving that MLAs are better at generalizing than traditional isotherm models. Pollutant adsorption on montmorillonite was assessed by Zhao et al. (2024a) using MLAs and Density Functional Theory (DFT) calculations. They concentrated on the gradient-boosting decision tree model and examined the factors influencing adsorption because it matched the experimental results. DFT advances our understanding of montmorillonite-based adsorbents by examining interactions at different pH levels through MLAs and theoretical calculations.

According to the literature review (Fanourgakis et al., 2019; Pardakhti et al., 2017; Yang et al., 2022), it can be concluded that the application of MLAs is popular for evaluating heavy metal adsorption processes using CDW. However, the integration of the RSM with MLAs and the subsequent design of control systems has not been well explored in previous studies, which will be addressed in the present research. Due to the complexity of adsorption processes, RSM can help assess key factors and predict adsorption performance, but it is not suitable for real-time operational management because of its reliance on predefined experimental designs and limited adaptability to continuous conditions (Anfar et al., 2020). Several studies have utilized RSM and hybrid modeling approaches in adsorption processes. Using Adaptive Neuro-Fuzzy Inference System (ANFIS) and RSM, Okoye et al. (2025) modeled the adsorption of Erythrosine B (EB) dye and achieved a satisfactory level of efficiency. Despite their excellent prediction capabilities, they failed to develop an alarm or decision-making system, which is the focus of this work. Also, Brendler et al. (2003) used Microsoft Access to create a relational sorption database (RES3T) based on Surface Complexation Models (SCMs), which allowed for the modeling and extraction of data relevant to individual minerals. However, this study addressed the lack of a predictive system. Moreover, Zafar et al. (2022) employed full-factorial central composite design-based batch trials to estimate chromium adsorption onto modified maghemite nanoparticles using ANN and ANFIS. They did not, however, create an early warning or control system, which is a topic covered in this study.

The present study aims to:

- Conduct experimental tests for the adsorption of Pb²⁺, Mn²⁺, and Co²⁺ ions onto Low Density Concrete (LDC) by pH, metal type, Initial Concentration (IC), adsorbent mass, and contact time.
- Assess initial data using descriptive statistical methods for data categorization.
- Apply the RSM for sensitivity analysis and process optimization.
- Utilize MLAs to enhance the adsorption process and design a soft-sensor for forecasting removal performance using various algorithms.
- Develop a decision-making model to control the process using Knowledge Flow (KF) platform in WEKA and MATLAB software.

By creating an expert system for the adsorption of heavy metals onto low-density concrete waste and combining real-time data analysis and control, this work introduces a significant innovation. In contrast to earlier research, the system is built with WEKA's KF platform, which facilitates predictive process management and dynamic decision-making. This invention supports intelligent water treatment in line with Industry 4.0 principles by bridging the gap between smart automation and experimental adsorption.

2. Methodology

The complete research plan is shown in Fig. 1, and each stage is briefly explained to clarify the study's procedure. Step 1 entails conducting laboratory adsorption tests to comprehensively assess the

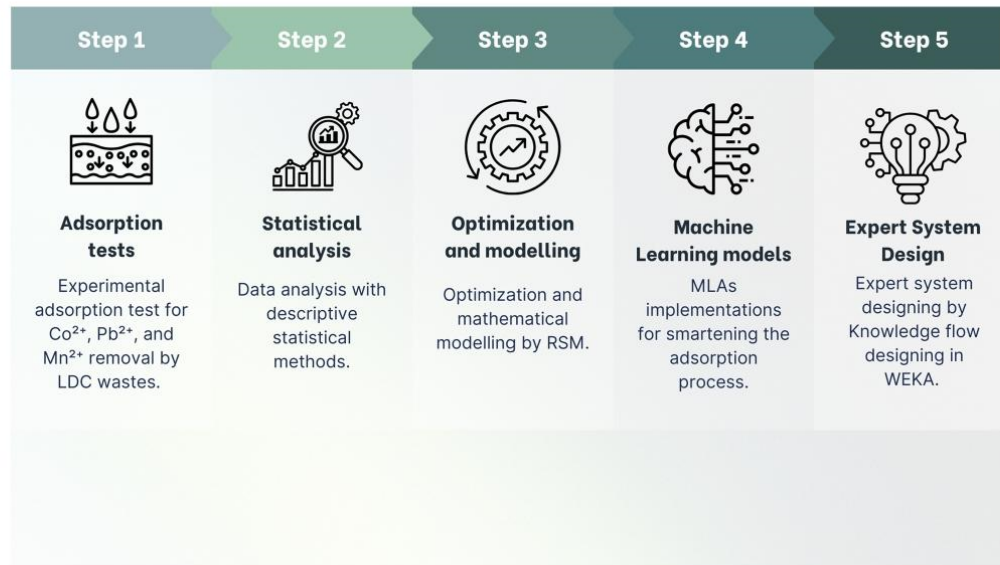


Fig. 1. The research roadmap of the present study.

removal behavior of Pb^{2+} , Mn^{2+} , and Co^{2+} ions onto LDC. The Removal Percentages (RPs) that arise from adjusting each of the five operational variables: pH, metal type, IC, adsorbent mass, and contact time, are monitored as the system output during this phase. In order to find preliminary trends and possible parameter interactions, the gathered experimental data are categorized and represented in Step 2's descriptive statistical analysis. Step 3 employs RSM to conduct mathematical modeling, optimization, and sensitivity analysis utilizing historical data. Although it is not highly flexible for real-time process control, this phase quantifies the impact of each operational parameter on RP and provides preliminary prediction information. To develop more precise soft-sensor models that can capture nonlinear correlations between the five input parameters and RP, Step 4 expands the analysis by applying 20 MLAs (ESI Table 1). To find the most trustworthy predictors, these MLAs are thoroughly assessed using statistical and categorization markers.

Finally, Step 5 utilizes WEKA's Knowledge Flow (KF) platform to integrate the optimized machine learning models into an expert-based Decision Support System (DSS). In order to forecast RP, categorize performance levels, and produce alarm-based operational guidance for intelligent water treatment management, real-time data streams are handled in this final stage.

The selection of pH, metal type, initial concentration, adsorbent mass, and contact time as input variables for modeling heavy metal adsorption is well-grounded in adsorption theory. pH is a dominant factor because it controls both the adsorbent surface charge and the chemical speciation of metal ions through protonation-deprotonation mechanisms, thus determining electrostatic interactions and possible

metal precipitation. Initial concentration provides the mass-transfer driving force and influences adsorption kinetics and equilibrium capacity. Adsorbent mass dictates the number of available active sites, although its effect may become non-linear due to aggregation or site saturation. Contact time reflects the kinetic progression of adsorption and indicates whether the process is diffusion- or reaction-controlled. Metal type is also crucial because ionic characteristics such as radius, hydration energy, and coordination behavior govern the affinity of metals for the adsorbent. Together, these variables capture the essential thermodynamic and kinetic properties of adsorption systems, making them necessary for accurate modeling and optimization (Raji et al., 2023). Therefore, these parameters were selected as inputs for the models in the present study.

2.1. Adsorption process

A produced CDW-derived adsorbent was used in the investigation by Gheibi et al. (2025) to decontaminate under various settings. These conditions included variable pH, IC of the metal ions, adsorbent mass, and time. All experiments were carried out at room temperature using the protocols provided by Gheibi et al. (2025). Following each experiment, which was carried out in a reactor with a volume of 15 mL, Eq. (1) was used to determine the RP:

$$RP(\%) = \frac{C_0 - C_e}{C_0} \times 100 \quad (1)$$

C_0 : Initial concentration (mg L^{-1})

C_e : Experimental concentration after adsorption (mg L^{-1})

The study systematically investigates how these factors affect the CDW adsorbent's adsorption effectiveness. The examined pH ranges that are examined span from acidic to approximately neutral, which is important since pH significantly impacts the metal oxidation state and the adsorbent's surface charge. The optimal amount required for maximal ion removal was determined by varying the adsorbent's mass. To understand the adsorption process kinetics and determine the time required to reach equilibrium, contact time was varied. Additionally, the initial metal ion concentrations were adjusted in order to assess the

Table 1
Ranges of input variables used in the modeling practices of this study.

Parameter	Unit	Min	Max
Heavy metal type	—	$\text{Pb}^{2+} = 1$, $\text{Co}^{2+} = 2$, and $\text{Mn}^{2+} = 3$	
pH	—	2	5
Initial concentration	mg L^{-1}	10	200
Adsorbent mass	g	0.02	0.15
Contact time	min	0	120

adsorbent's capacity and effectiveness at various contaminant concentrations.

In this research, using a One-Factor-At-a-Time (OFAT) approach, the study systematically varied pH, adsorbent mass and contact time to investigate the adsorption behavior of heavy metals. The present investigation used OFAT to isolate each factor's effect before interaction modeling as per the outcomes of Gheibi et al. (2025). Its 66 data points were then employed to support RSM-based analysis and subsequent MLAs modeling. The ranges of input data applied in this study are shown in Table 1. Experiments were carried out at an acidic pH range of 2.0 to 5.0. In the first step, 15 mL of a 10 mg L⁻¹ metal ion solution was mixed with 0.1 g of a fixed dose of LDC adsorbent, kept at 25 °C, and agitated at 400 rpm. Adsorbent mass ranged from 0.02 to 0.15 g, while contact time ranged from 0 to 120 min. Following the specified contact time, samples were spun for 10 min at 3000 rpm. Atomic Absorption Spectroscopy (AAS) was used to determine the residual concentrations of heavy metals using element-specific wavelengths for Pb²⁺, Co²⁺, and Mn²⁺. Each metal ion was then subjected to isothermal adsorption studies across a concentration range of 10–200 mg L⁻¹. For use in data mining and machine learning analysis, the whole dataset gathered from these tests was assembled. The schematic plan of the experiments conducted in this study is presented in ESI Fig. 1. ESI Fig. 1 illustrates the experimental workflow used in this study. Aqueous solutions containing Pb²⁺, Co²⁺, and Mn²⁺ ions were first prepared at controlled pH. A known mass of LDC powder was then added and mixed for a selected contact time to allow adsorption. After reaction, solid-liquid separation was performed using centrifugation, and the clarified supernatant was collected. The remaining metal concentrations were subsequently quantified by AAS, while the separated solid phase contained the adsorbed heavy metal ions (Gheibi et al., 2025).

2.2. Data analysis

The initial experimental data are presented using split heat map plots. These visualizations illustrate the effects of paired parameters against RP. This categorization allows for a preliminary sensitivity analysis of the input parameters. However, a more comprehensive sensitivity analysis will be conducted in subsequent steps using more precise statistical techniques. All analyses in this section were performed using OriginPro 2021 software.

2.3. RSM technique

In the present study, historical data analysis as part of the RSM method in Design Expert 7.0.0 is applied. The mathematical model considers five input factors: pH, metal type, IC, adsorbent mass, and time, with RP as the output. In the RSM analysis, curve fitting is first performed to identify the best equation for predicting RP. Then, ANOVA analysis is conducted to determine the most significant factors. Finally, after a sensitivity analysis of the key features in the experiments, the optimization process is performed. The general form of the mathematical equation in the curve-fitting process is demonstrated in Eq. (2) (Mahmoudi Jalali et al., 2023).

$$Z = a_0 + a_1X_1 + a_2X_2 + \dots + a_nX_n + a_{12}X_1X_2 + a_{13}X_1X_3 + \dots + a_{1n}X_1X_n + \dots + a_{(n-1)n}X_{n-1}X_n + a_{11}X_1^2 + \dots + a_{nn}X_n^2 \quad (2)$$

2.4. MLAs

In the next stage of the study, 20 algorithms in WEKA software version 3.9.6, including Function Gaussian Process (FGP), Function Linear Regression (FLR), Function Multilayer Perceptron (FMLP), Function Simple Linear Regression (FSLR), Function SMOreg (F SMOreg), lazy Instance-Based K (IBK), lazy KStar, lazy Locally Weighted

Learning (LWL), Meta Additive Regression (MAR), Meta Bagging (MB), Meta Random Committee (MRC), Meta Randomizable Filtered Classifier (MRFC), Meta Random Sub Space (MRSS), Meta Regression by Discretization (MRD), Rules Decision Table (RDT), Rules M5 Rules (RM5R), Trees Decision Stump (TDS), Trees M5P (TM5P), Trees Random Forest (TRF), and Trees Random Tree (TRT) (Hall and Reutemann., 2025). The descriptions of all the applied algorithms are presented in ESI Table 1. In the first step, all the algorithms are compared for the adsorption data analysis in 60 % Split Percentage (SP) of training and 40 % for testing. In machine learning, the SP describes splitting a dataset into discrete portions, usually for testing and training (Hall and Reutemann., 2025). The 60/40 split was initially selected to perform a stricter early-stage screening of the 20 machine-learning algorithms. Using a larger test portion (40 %) increases the evaluation pressure on each model, helping identify only the most robust algorithms under limited training data. After this screening, the selected models were further optimized, and an 80/20 split was later applied to maximize training data and achieve the highest predictive accuracy.

Then, the results are compared together according to statistical indicators, including Correlation Coefficient (CC), Mean Absolute Error (MAE), Root Mean Squared Error (RMSE), Relative Absolute Error (RAE), and Root Relative Squared Error (RRSE) (Horn, 1993). All mathematical expressions of the indicators are presented in ESI Table 2. In this stage, the algorithms will be screened, and the ones with CC > 0.92 will be selected. In the next step, due to the evaluation of selected algorithms, some other classification indicators (ESI Table 3), including True Positive Rate (TP Rate), False Positive Rate (FP Rate), Precision, Recall, F-measure, Matthews Correlation Coefficient (MCC), Receiver Operating Characteristic (ROC) Area, and Precision-Recall Curve (PRC) Area (Poh et al., 2018). The algorithms classified samples into two classes (C₀ and C₁) derived from the continuous RP removal values using model-specific, data-driven decision rules in WEKA software. Standard classification metrics were then employed solely to benchmark the performance of different MLAs. Finally, the most accurate algorithms, as per statistical and classification indicators, will be tuned using hyperparameter optimization. Plus, it should be noted that the application of multiple machine learning algorithms (Liu et al., 2024) can serve as a promising approach for controlling complex processes under unpredictable conditions. For instance, in cases involving unknown waste materials, this multidimensional technique can enhance the system's adaptability and overall intelligence.

2.5. Expert system design

Further, the optimal performance of MLAs is implemented as an expert system with a knowledge flow package of WEKA software. In the analysis, a system is developed for online data mining of adsorption processes in water treatment facilities, including urban water treatment plants and mine drainage treatment systems. All data analysis and categorization plus mining of them are executed by KF toolboxes. Finally, visualization and reporting tools are arranged there (Hall and

Table 2

The results of ANOVA assessment in the present study.

Source	Sum of Squares	Df	Mean Square	F-Value	p-value	
Model	33,730.54	5	6746.1	69.12	< 0.0001	significant
A-Metal type	15,852.18	1	15,852.18	162.4	< 0.0001	
B-pH	12,582.62	1	12,582.62	128.91	< 0.0001	
C-M	5371.36	1	5371.36	55.03	< 0.0001	
D-Time	332.39	1	332.39	3.4	0.07	
E-IC	396.54	1	396.54	4.06	0.05	

Table 3

The optimal suggestions for effective factors in the adsorption of heavy metals onto LDC.

Metal type	pH	M (g)	Time (min)	IC (mg L ⁻¹)	RP (%)
Pb ²⁺	5	0.13	33.5	35	99
Co ²⁺	5	0.15	110	10	99
Mn ²⁺	5	0.15	120	10	81

Reutemann., 2025).

After designing the KF, the model was further extended into a control dashboard using MATLAB 2019b. The initial assessments were conducted in WEKA; however, to enhance dynamism, flexibility, and implementation capability, the model was subsequently redesigned and developed in the MATLAB environment. The model architecture begins with a CSV Loader that imports experimental data, followed by an RP Class Assigner that categorizes RP values into four predefined classes including [11 %–33 %], [33 %–55 %], [55 %–77 %], and [77 %–99 %]. A Fold Maker then partitions the dataset into training (70 %) and testing (30 %) subsets. A baseline MLA is trained first, after which a Sensitivity Analyzer systematically evaluates key RF hyperparameters to identify the optimal configuration. The optimized MLA model performs continuous RP prediction, which is further translated into discrete RP classes for decision-making. These predictions feed into an Alarm Manager that detects low-RP conditions and triggers alerts. Finally, a comprehensive

Dashboard visualizes performance metrics, class distributions, and sensitivity outcomes, while a Results Exporter saves all generated reports and prediction tables.

3. Results and discussions

In this section, the experimental data were first analyzed using statistical methods to evaluate variations under different experimental conditions. The primary objective was to identify patterns and fluctuations in the data. In the next phase, RSM and sensitivity analysis were applied to determine the optimal operating conditions. However, the resulting model at this stage was not suitable for real-time prediction and dynamic control of the heavy metal adsorption process. Therefore, in the following step, machine learning models were implemented for both regression (prediction) and classification (screening of removal percentages) tasks. Finally, the developed model was integrated into a dynamic system using the KF concept to serve as an expert-based, real-time warning system for adsorption processes.

3.1. Experiments and data analysis

A set of heat maps showing the experimental association between RP and other effective factors for heavy metals treatment is shown in Fig. 2 (a-d). Before RSM and machine learning models were applied, heat maps were utilized as an initial sensitivity analysis to visually identify

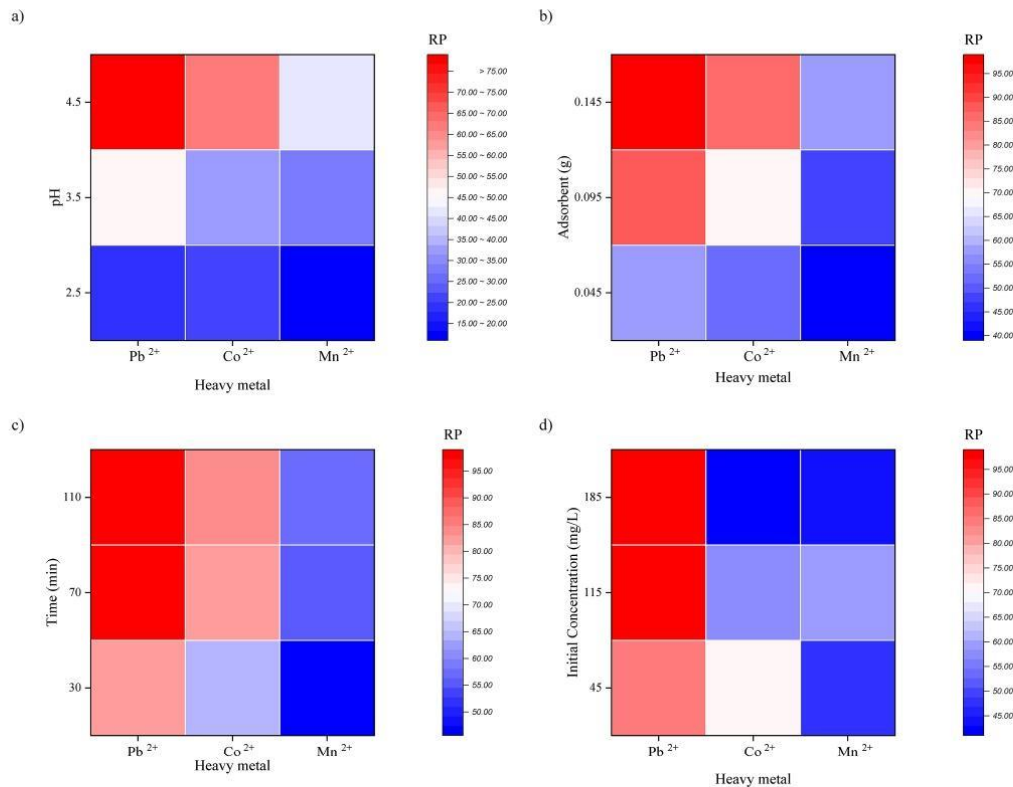


Fig. 2. The heat map of data categorization of adsorption parameter for different metal types (Pb²⁺, Co²⁺, and Mn²⁺) vs. RP for (a) pH, 2 < pH < 5, (b) adsorbent amount, 0.02 g < adsorbent (g) < 0.15 g, (c) time, 10 min < Time (min) < 120 min, (d) initial concentration, 10 mg L⁻¹ < concentration (mg L⁻¹) < 200 mg L⁻¹. General conditions are pH = 5, IC = 10 mg L⁻¹ of each heavy metals, mass of adsorbent = 0.1 g of LDC powder, contact time = 30 min, and solution volume = 15 mL.

parameter interactions and early trends in adsorption behavior. This phase made it possible to identify the variables that have the most significant effect, especially the significant impact of pH on removal performance. It also made it easier to identify nonlinear or metal-specific trends that are not visible from raw data alone. Pb^{2+} , Co^{2+} , and Mn^{2+} specific trends have been clearly presented in the Results section, which has been updated to highlight the experimentally derived results.

Fig. 2(a) shows how pH affects the RP of various heavy metals with values ranging from 2 to 5. Based on heavy metals solubility product constant (K_{sp}), the pH range of 2–5 was analyzed. Because Pb^{2+} , Co^{2+} , and Mn^{2+} at a concentration of 200 mg L⁻¹ precipitate at pH of ~ 7.5, 7.79, and 8.82, respectively, this concentration was taken as the highest used in experiments. As pH increases above 5, ions form metal hydroxide precipitates ($M(OH)_2$), reducing adsorption effectiveness (Harris, 2010). To prevent precipitation, a maximum pH of 5 was selected. The adsorption of all heavy metals increased as pH increased. Although pH is the main driver, sensitivity analysis provides a more nuanced assessment of other factors, like IC and adsorbent mass, to better understand their respective contributions. To ascertain if a solid will precipitate from a solution, precipitation control calculations use the K_{sp} . The K_{sp} formulation is $K_{sp} = [A^+]^m[B^{n-}]^n$ for a salt A_mB_n , which dissociates as $A_mB_n \rightleftharpoons mA^{m+} + nB^{n-}$. The present study compares the ion product (Q) to K_{sp} by computing it using the real ion concentrations in solution. A precipitate occurs and the solution is supersaturated if $Q > K_{sp}$. There is no precipitation if $Q \leq K_{sp}$. With the help of this technique, precipitation in chemical reactions can be precisely predicted and controlled, guaranteeing stability or causing solid formation as necessary (Akitsu, 2017; Clifford, 2002).

Fig. 2(b) shows dependency of adsorbed amounts to amount of sorbent used (solid/liquid ration). Adsorbed amount is increasing with amount of adsorbent, which is logical. Fig. 2(c) presents the impact of metal ion concentration on RP at different times, showing a dynamic interplay between concentration and time. These findings align with previous studies that emphasize the role of pH in optimizing adsorption efficiency, as discussed below. The same results on the effects of time-pH are evaluated in the adsorption process by Chen et al. (2011). They demonstrated that the adsorption of Cd^{2+} on a type of bentonite achieves equilibrium in 50 min, with increasing pH enhancing adsorption capacity and specific pH levels and contact times significantly influencing the adsorption process (Chen et al., 2011). Likewise, Jamaloi et al. (2025) demonstrated that increasing the pH during the adsorption of Co^{2+} ions using bone powder modified with polypyrrole and polyethylene glycol enhances the removal efficiency. Plus, Pooladi et al. (2024) reported that increasing the pH from 2 to 6 improved the adsorption performance of Pb^{2+} onto a biosorbent derived from Cardita bicolor shell. Similarly, Zadeh & Jafari (2024) demonstrated that increasing the pH from 2 to 6 significantly improves the adsorption efficiency of Pb^{2+} onto an activated carbon/alginate/ Fe_3O_4 magnetic nanocomposite.

According to Fig. 2(d), with increasing IC, the percentage of IC adsorbed is increasing in Pb^{2+} and reversely in Co^{2+} . While, in Mn^{2+} , the fluctuations was high and it shows low correlation between IC and RP in the case of Mn^{2+} and therefore, the effects of other features should be evaluated in multidimensional assessment. Notable variations in RP imply that pH is an important factor in metal adsorption. This uncertainty in Mn^{2+} adsorption patterns with IC emphasizes the need for multidimensional analysis that takes into account several factors at once, including pH and contact time. However, as Fig. 2(b–d) demonstrates, the RP is also influenced by other variables, such as time, IC, and adsorbent mass. Since variations in the heat map are more noticeable when pH is altered than when other parameters are considered, this implies that pH might be one of the most influential factor in the adsorption of heavy metals from water supplies by LDC. This result is in line with earlier research conducted by Kavand et al. (2014) and Xu et al. (2013). However, this research needs more validation utilizing exact methodologies, especially taking into account the pH changes during the

adsorption process (not just using the initial pH value prior to adsorption).

As observed in the present study, LDC exhibits higher efficiency in the decontamination of Pb^{2+} compared to Co^{2+} and Mn^{2+} . Similarly, Nemati et al. (2021) reported that Pb^{2+} demonstrated superior adsorption performance over Ag^{3+} when using activated carbon derived from Citrus limon tree leaves. Furthermore, Aboli et al. (2020) showed that Pb^{2+} outperformed Co^{2+} and Ni^{2+} ions in adsorption onto activated carbon prepared from Citrus limetta leaves. Lastly, Koozhad et al. (2019) confirmed that Pb^{2+} had a higher adsorption efficiency than As^{3+} when using activated carbon produced from Tamarix leaves. These similar findings across multiple studies further support the validity of the current results.

3.2. Optimization and sensitive analysis

Curve fitting using RSM was used to further validate the experimental patterns seen in Fig. 2, and the outcomes are compiled in ESI Table 4. The linear model recommended by the RSM technique has an R^2 value of 0.85 and a predicted R^2 of 0.80, as shown in ESI Table 4. According to these findings, 85 % of the variability in the response variable can be explained by the linear model, and the anticipated R^2 value of 0.80 indicates a respectable degree of predictive power. Regression coefficients of 0.80 are deemed insufficiently dependable for precise prediction in industrial applications. This suggests that more reliable prediction models are required. Because machine learning algorithms can handle complicated, non-linear interactions in data, they may, therefore, be a good alternative for improving adsorption performance prediction. The obtained equation for the RSM technique is demonstrated in Eq. (3). According to the formula, it can be understood that with different values of metal type (Pb^{2+} : 1, Co^{2+} : 2, and Mn^{2+} : 3), the RP could be calculated in different pH, IC, adsorbent mass (M), and time. The coefficients of different factors are different because of their different scales and units. Similarly, Urréjola-Madrinán et al. (2022) proposed a linear model to describe the adsorption of Cr(VI) using spirulina algae biomass. In their study, they characterized the adsorption capacity using a linear relationship involving the IC and the mass of the adsorbent.

$$RP(\%) = -19.69 + (-18.98 \times Metaltype) + (18.83 \times pH) + (360.49 \times M) + (0.08 \times Time) + (-0.05 \times IC) \quad (3)$$

According to ESI Fig. 2(a), it is clear that the experimental RP is distributed around the normal diagram, and therefore, the model follows the normal statistical distribution. Also, in the study of Ahmaddadeh & Hemmati (2025), the response of Pb^{2+} adsorption onto zinc-based MOFs-derived carbon indicates that the residuals are normally distributed. As shown in ESI Fig. 2(b), standardized residuals with values between -3 and 3 indicate that most data points are in a line with traditional regression analysis predictions, implying a generally normal distribution and adequate model. Anything outside of this range might need further examination for abnormalities in the data and model validity. Likewise, analysis of ESI Fig. 2(c) reveals a close alignment between actual and predicted values, showing good fit and random distribution around the regression line. At the same time, there are some differences between the actual and prediction amounts. This difference indicates potential for further model refinement to enhance predictive accuracy.

Lambda values in ESI Fig. 2(d) from the Box-Cox diagram help determine the best data transformation for model accuracy. The lambda values are: Current = 1, Best = 0.73, High = 1.22, and Low = 0.3. This means that the current transformation (lambda = 1) works well enough, but adjusting it to 0.73 could improve model accuracy. The range (0.3 to 1.22) shows how sensitive the model is to the lambda value, which can affect data normalization quality. Since the current lambda provides good results, no further transformation is needed (Sreeharsha et al.,

Table 4
Summary of various studies on the application of MLAs in adsorption processes.

Adsorbent	Adsorbate	Machine learning model	Statistical indicators	Reference
Metal-organic frameworks	As ^{3+/5+} , Cd ²⁺ , Cr ⁶⁺ , Cu ²⁺ , Pb ²⁺ , Hg ²⁺	Gradient Boosting Decision Tree (GBDT)	R ² =0.921-0.962 for 6 heavy metals	(Wang et al., 2025)
Biochar	Cu ²⁺ , Zn ²⁺ , Pb ²⁺ , Cd ²⁺ , Ni ²⁺ , As ³⁺	Random forest	R ² = 0.973	(Zhu et al., 2019)
Bentonite	Pb ²⁺ , Cu ²⁺ , Cd ²⁺ , Zn ²⁺ , Ni ²⁺ , Co ²⁺ , and chromium ions	eXtreme Gradient Boosting Regression (XGB)	Train R ² = 0.995 Test R ² = 0.9359	(Guo et al., 2024)
Hydrochar	Pb ²⁺ , Cd ²⁺ , Zn ²⁺ , Cu ²⁺ , Sb ³⁺ , Sb ⁵⁺ , CrO ₄ ²⁻ , UO ₂ ²⁺	Gradient boosting decision tree	R ² = 0.93-0.98	(Zhao et al., 2023)
Clay minerals	Cd ²⁺ , Cu ²⁺ , Pb ²⁺ , Zn ²⁺ , Ni ²⁺ and Co ²⁺	Extreme Gradient Boosting	R ² = 0.977	(Zhao et al., 2024b)
LDC	Pb ²⁺ , Co ²⁺ , Mn ²⁺	Trees Random Forest	R ² = 0.98	This study

2020). Therefore, the existing condition is used for modeling.

As we discussed about our normal distribution of results, also, Danmaliki et al. (2017) showed that the adsorption process in the desulfurization on nickel/activated carbon follows a normal distribution. A similar conclusion had Saini et al. (2019), who studied Cd²⁺ adsorption onto C_{16.6-16} incorporated mesoporous MCM-41. This will be further elaborated in the following sections.

The results of the Analysis of Variance (ANOVA) are presented in Table 2. According to the table, the p-value of the model is <0.0001, indicating that the model is highly significant. Among all the influential factors, pH, metal ion type and mass of adsorbent exhibit significant effects on RP with p-values < 0.0001. Specifically, the most substantial

influences are attributed to metal ion type (162.4) and pH (128.91). In contrast, the impacts of IC and reaction time on RP are relatively minimal. The reason for these is that relative narrow intervals of values for IC and reaction time was studied in the experimental setup.

Sensitivity analysis was performed to visualize the interactions and comparative influence of pH and metal ion type, which were identified as dominant components in the ANOVA results. The sensitivity analysis results are displayed in Fig. 3. The adsorption process efficiency of Pb²⁺, Co²⁺, and Mn²⁺ varies significantly. These variations can be related to variances in their ionic radii. In particular, it is clear that Pb²⁺ has higher adsorption effectiveness than both Co²⁺ and Mn²⁺, indicating that ionic radius has an effect on the adsorption mechanism. Also, Dong

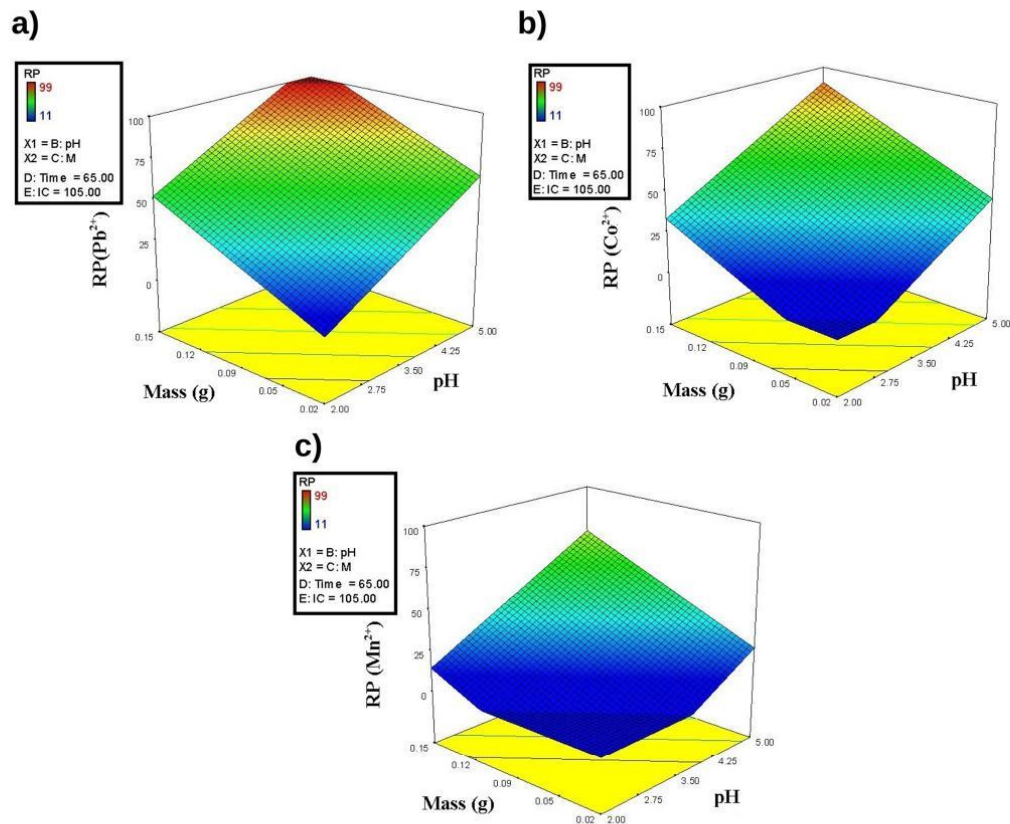


Fig. 3. The sensitive analysis of pH vs. mass of adsorbent as per (a) Pb²⁺ RP, (b) Co²⁺ RP, and (c) Mn²⁺ RP.

et al. (2007) conducted research demonstrating that the adsorption capacity of Pb^{2+} exceeds that of Co^{2+} on various metal oxides and organic materials found in natural surface coatings. Their findings corroborate the results observed in the present study. This underscores the consistent pattern of Pb^{2+} exhibiting higher adsorption capacity compared to Co^{2+} across different materials studied in both investigations.

The differences in maximum RP values seen among various metal ions are directly caused by these ideal parameter combinations. The optimal conditions for adsorbing Pb^{2+} , Mn^{2+} , and Co^{2+} ions onto LDC are summarized in Table 3. According to the table, the ideal pH and mass of the adsorbent are consistently 5 and 0.13–0.15 (ESI Fig. 3a–c), respectively, for all ions. Specifically, LDC demonstrates remarkable efficacy for Pb^{2+} ions, three times higher IC and reaching maximum adsorption capacity in three times less contact time compared to Mn^{2+} and Co^{2+} ions.

In the following, based on the model, the maximum adsorption efficiency for Co^{2+} ions can reach up to 99 %, whereas for Mn^{2+} ions, it reaches 81 % (ESI Fig. 3b and c). These results underscore the varying adsorption capacities of LDC for different metal ions, reflecting its potential as an effective adsorbent in environmental remediation processes. Similar to the results of our study, Çelebi et al., 2020

demonstrated that the optimal removal efficiency for Zn^{2+} and Ni^{2+} ions occurs at a pH of approximately 5 during their experiments. Moreover, Meena et al. (2024) demonstrated that As^{3+} adsorption onto cross-linked chitosan–tripolyphosphate nanoparticles achieves optimal capacity at approximately pH 5. Plus, as per the study of Nyirenda et al. (2022), the maximum adsorption of Cu^{2+} , Pb^{2+} , Cd^{2+} , and Zn^{2+} ions onto the silver–silica nanocomposite supported by activated carbon occurred at approximately pH 5.5, aligning with the findings of the current study.

The RSM model's structure is intrinsically static and cannot self-train, update, or adapt when new data or operational disruptions arise, despite the fact that it attained respectable statistical accuracy. RSM is primarily appropriate for mechanistic interpretation and sensitivity analysis rather than real-time automation because it depends on predetermined experimental designs and established polynomial equations. The current study, on the other hand, calls for a dynamic predictive engine that can support automated control actions and continually learn from incoming sensor data. By recording nonlinear behaviors, retraining with fresh data, and integrating straight into the KF platform, where real-time prediction, categorization, and alarm management are implemented MLAs overcome the limitations of RSM. Therefore, MLAs are chosen not only to increase accuracy but also to enable an intelligent system that is

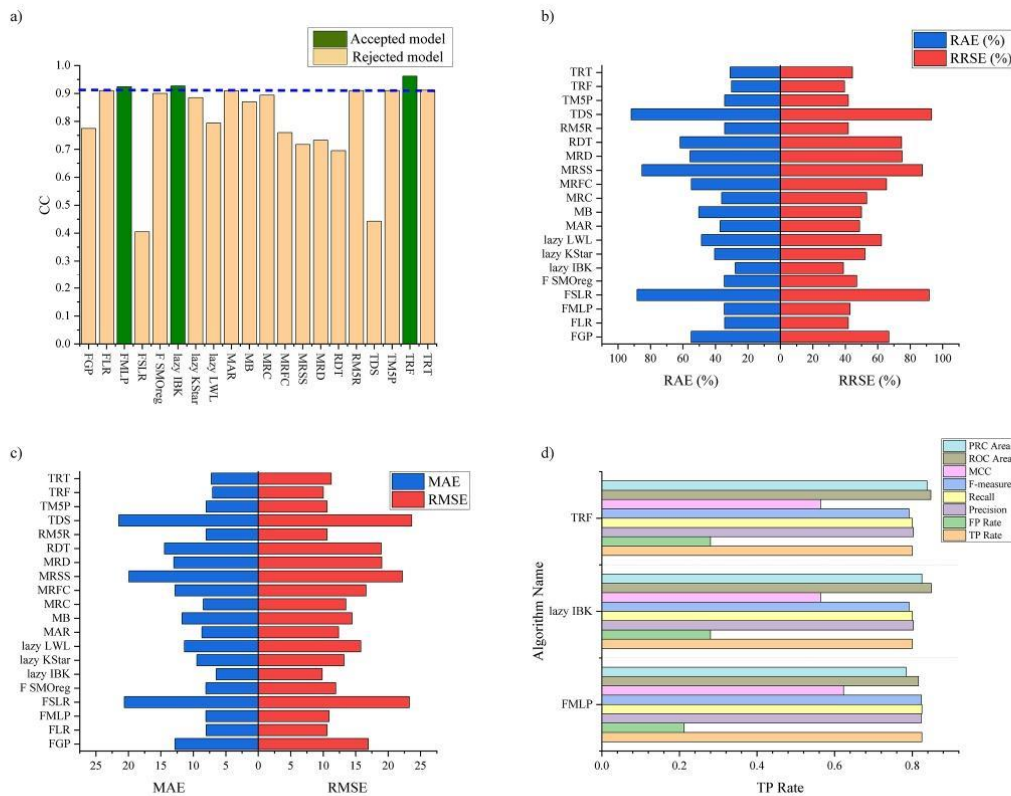


Fig. 4. The outputs of machine learning analysis based on (a) CC of 20 algorithms and categorizations, (b) RAE % and RRSE % for 20 algorithms, (c) MAE and RMSE indicators for 20 algorithms, and (d) classification performance metrics in three selected algorithms. Abbreviations - CC: Correlation Coefficient, MAE: Mean Absolute Error, RMSE: Root Mean Squared Error, RAE: Relative Absolute Error, and RRSE: Root Relative Squared Error, FGP: Function Gaussian Process, FGLR: Function Linear Regression, FMLP: Function Multilayer Perceptron, FSLR: Function Simple Linear Regression, F SMOreg: Function SMOreg, IBK: Lazy Instance-Based K, KStar: Lazy KStar, LWL: Lazy Locally Weighted Learning, MAR: Meta Additive Regression, MB: Meta Bagging, MRC: Meta Random Committee, MRFC: Meta Randomizable Filtered Classifier, MRSS: Meta Random Sub Space, MRD: Meta Regression by Discretization, RDT: Rules Decision Table, RM5R: Rules M5 Rules, TDS: Trees Decision Stump, TMSP: Trees MSP, TRF: Trees Random Forest, TRT: Trees Random Tree.

compatible with Industry 4.0, something that RSM is essentially unable to achieve.

3.3. Machine learning algorithms

Previously, we modeled the RP values based on several effective factors, including pH, metal type, IC, adsorbent mass, and time in the solution. We also conducted a sensitivity analysis to determine the most significant factors affecting RP among these parameters. Initially, we developed a regression-based model, which, however, required frequent adjustments. MLAs can overcome this limitation by enabling automated training processes based on input data at specific intervals. A promising substitute for more accurate prediction is provided by machine learning algorithms, as the linear regression model is unable to adequately capture intricate nonlinear interactions.

In the following sections, various MLAs were explored to predict RP based on these effective features as dynamic soft-sensors. Both statistical and classification indicators were examined for the different algorithms, and the selected MLAs were tuned accordingly. The results of the MLA analysis are shown in Fig. 4 and ESI Table 5. According to Fig. 4(a), it is evident that three algorithms, FMLP, lazy IBK, and TRF, exhibit a CC exceeding 0.92. These algorithms are considered the best performers at this stage, having been trained with 60 % of the data. A correlation coefficient above 0.92 indicates a strong relationship between the predicted (by the mentioned algorithms) and actual values (experimental results) in a machine learning model. Physically, this means the model's predictions closely match the water treatment processes through adsorption technique, suggesting high predictive accuracy. It implies that changes in the input variables are strongly linked to the changes in the target variable, allowing the model to make reliable predictions (Eftekhari et al., 2021).

According to Fig. 4(b), when examining the RAE %, which measures the total absolute error in relation to a simple predictor, lazy IBK achieves the lowest RAE % at 27.73, followed by TRF at 30.11. Figs. 4 (a-c) indicate that these models have a lower proportion of error relative to a naive benchmark model. TDS, with a RAE % of 91.91, shows the highest relative error, further indicating its inefficiency compared to simpler models. In Fig. 4(b), the RRSE %, which normalizes the RMSE by the standard deviation of the actual values, is lowest for lazy IBK (38.91) and TRF (39.54), suggesting these models have a smaller proportion of squared error relative to other models. TDS, with an RRSE % of 93.22, again has the highest value, emphasizing its poor performance. Regarding the MAE indicator in Fig. 4(c), which indicates the average magnitude of the errors in a set of predictions, lazy IBK stands out with the lowest MAE of 6.48, followed by TRF at 7.03. These low values imply that, on average, the predictions made by these models are very close to the actual values. Conversely, TDS has the highest MAE at 21.47, indicating larger average prediction errors and lower accuracy. The RMSE, which provides an indication (Fig. 4c) of the model's error magnitude and is more sensitive to outliers, is also lowest for lazy IBK (9.86) and TRF (10.02). These low RMSE values highlight the models' robust performance in minimizing error. TDS, with the highest RMSE of 23.64, again demonstrates the poorest performance among the algorithms in this aspect.

Finally, TRF and lazy IBK emerge as the top performers across various metrics. TRF achieves the highest correlation coefficient and low error rates, establishing it as a reliable and accurate model. Lazy IBK demonstrates excellent performance with the lowest MAE and RMSE, highlighting its high predictive accuracy and robustness. Additionally, FMLP exhibits commendable efficiency following lazy IBK and TRF, achieving approximately 0.92 CC.

In a study by Yaqub et al. (2020), it was demonstrated that an ANN with an R^2 value of 0.97 is the most effective technique for predicting the removal efficiency of Ca^{6+} purification using polymer inclusion membranes. The origin of ANN is similar to that of the FMLP. In the present research, FMLP has also been identified as one of the best algorithms for

estimating adsorption efficiency. Interestingly, in a study by Hafsa et al. (2021), the CC was calculated to be approximately 0.97 using the TRF algorithm for predicting arsenic removal from water resources. This result is comparable to the findings of the present research regarding the TRF algorithm.

Moreover, Jaskulak et al. (2020) compared the performance of ANN with RSM for predicting Cd^{2+} removal efficiency by *Sinapis alba* L. Similar to the present study, their analysis demonstrated that ANN, specifically with the Levenberg-Marquardt algorithm, achieved a CC of approximately 0.99, which was higher than that of RSM.

In the present study, it has been proven that TRF, lazy IBK, and FMLP algorithms exhibit higher accuracy in estimating RP compared to RSM. However, it is noteworthy that the ANN algorithm involves extensive computational stages. At an industrial level, utilizing algorithms such as TRF or lazy IBK can significantly reduce prediction time and investment costs in smart infrastructure development.

Fig. 4(d) shows that FMLP outperforms lazy IBK and TRF in key areas. FMLP has a higher TP Rate (0.825) than lazy IBK and TRF (0.80), indicating better accuracy in detecting real positives. Additionally, FMLP has a lower FP Rate (0.21) compared to lazy IBK and TRF (0.28), resulting in fewer false positives and increased reliability.

FMLP performs slightly better than lazy IBK and TRF in precision (FMLP: 0.80) and recall (FMLP: 0.82). This means FMLP is more accurate and reliable in predicting positives, making it preferable in applications where precision and recall are crucial. Also, the same results are obtained from F-measure and MCC indicators analysis.

In terms of ROC Area, which measures the ability of the classifier to distinguish between classes, lazy IBK and TRF show similar performance with scores of 0.84 and 0.84, respectively, slightly higher than FMLP's 0.81. However, PRC Area, which focuses on precision and recall across thresholds, highlights TRF's superiority with the highest score of 0.83, followed by lazy IBK at 0.82 and FMLP at 0.78. Therefore, in metrics directly related to classification accuracy, such as TP Rate, FP Rate, Precision, Recall, F-measure, and MCC, FMLP generally performs better than lazy IBK and TRF. Lazy IBK and TRF, on the other hand, perform marginally better in the ROC and PRC regions; TRF has the highest PRC area, indicating superior precision-recall performance across various thresholds. The exact application requirements ultimately determine which approach is best, especially whether improved ROC and PRC performance or increased precision and recall are more important. Based on engineering judgment and operational requirements, these three algorithms should operate simultaneously on the dashboard, allowing operators to decide their use based on experience.

Al-Sakkari et al. (2023) conducted an impressive study in which they used decision tree classification models and machine learning models to estimate the efficiency of adsorption-based carbon dioxide capture materials. Their findings attained recall, precision, and F-measure values of 1. The performance of the models in this study varied due to differences in data sources and experimental setups. Yang et al. (2022) applied a decision tree classification model for acetylene adsorption by MOFs. They found that the model achieved recall, precision, and F-measure values of 86.40, 92.0, and 88.80, respectively. The recall and F-measure values reported by Yang et al. (2022) are similar to the results of the present study.

The impact of varying the SP on the operation of the selected algorithms (FMLP, lazy IBK, and TRF) can be analyzed through statistical metrics to understand their robustness and trustworthiness under different conditions (Fig. 5).

According to Fig. 5(a), TRF steadily achieves the highest CC, reaching up to 0.99 at SP=80 %, demonstrating a very strong linear relationship regardless of the SP. FMLP and lazy IBK also illustrate large CC amounts, with FMLP peaking around 0.95 at SP=65 % and SP=80 %, and lazy IBK reaching 0.96 at SP=75 %. However, TRF generally outperforms the other two algorithms in terms of CC, suggesting superior reliability in its predictions.

In consideration of Fig. 5(b), MAE indicates the average magnitude

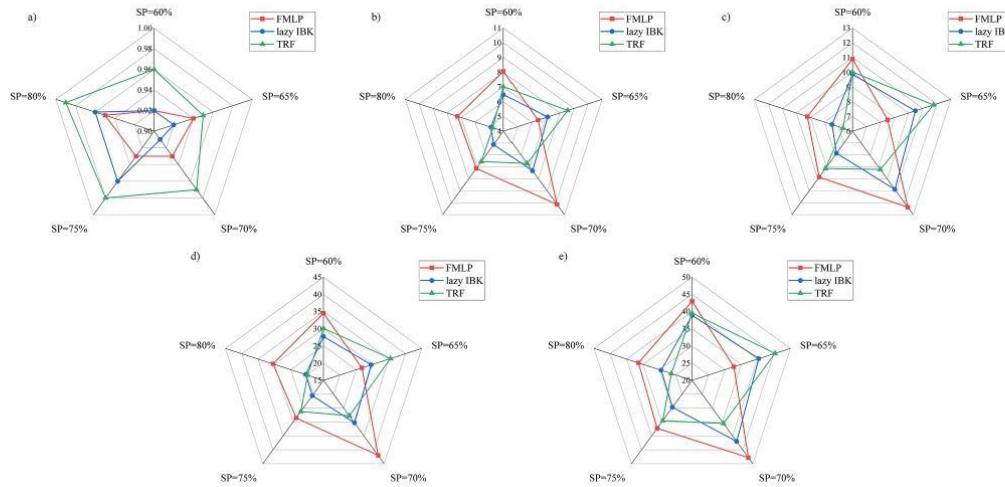


Fig. 5. The results of machine learning analysis in different SP % = 60 %–80 % based on (a) CC, (b) MAE, (c) RMSE, (d) RAE % and (e) RRSE %.

of errors in the predictions. At SP = 60 %, lazy IBK has the lowest MAE (6.48), followed by TRF (7.03) and FMLP (8.06). As SP increases to 80 %, TRF shows significant improvement, achieving the lowest MAE (4.75), indicating enhanced accuracy with a larger training set. Lazy IBK also shows improvement, with an MAE of 4.88 at SP = 80 %, while FMLP's MAE decreases to 7.28. TRF and lazy IBK benefit more from larger training sets than FMLP, with TRF showing superior performance. Lazy IBK also improves significantly with more data, while FMLP generally achieves acceptable results, especially at higher split percentages. This highlights the importance of tuning and larger training sets for algorithm performance.

According to Fig. 5(c) and d, at SP 60 %, lazy IBK has the lowest RMSE (9.86), TRF follows (10.02). At SP 80 %, both improve (lazy IBK: 7.46, TRF: 6.68). RAE at SP 60 %, lazy IBK leads (27.73 %); at SP 80 %, TRF improves (19.87 %), surpassing lazy IBK (20.43 %). RRSE at SP 60 %, lazy IBK is lowest (38.91 %); at SP 80 %, TRF is best (26.35 %). TRF and lazy IBK benefit more from larger training sets than FMLP. As mentioned before, finally, the present study applied TRF for creating the expert system because of its superior performance in precision-recall metrics, particularly in achieving the highest PRC area, which indicates its effectiveness across varying decision thresholds. TRF's consistent reliability in these aspects made it the preferred choice for developing the expert system, aligning with the operational requirements and practical application needs of the system.

Table 4 shows that MLAs such as Gradient Boosting Decision Tree (GBDT) (Wang et al., 2025), Random Forest (Zhu et al., 2019), and XGBoost (Guo et al., 2024) achieve high accuracy ($R^2 \approx 0.92$ – 0.99) across diverse adsorbents, indicating their strong ability to capture complex adsorption behavior. However, the results also suggest that the surface chemistry and heterogeneity of each adsorbent strongly influence which algorithm performs best. Materials like biochar, bentonite, and hydrochar-rich (Zhao et al., 2023) in functional groups and porous structures align well with tree-based models capable of handling nonlinear adsorption patterns. The present study similarly shows that LDC, with its heterogeneous $\text{CaO-SiO}_2\text{-Al}_2\text{O}_3$ surface, is best modeled by TRF ($R^2 = 0.98$). This highlights a key future direction: identifying which surface-adsorbate interaction patterns correspond to specific MLA architectures, enabling more targeted and mechanistic model selection in adsorption research.

3.4. Knowledge flow system design

The next obvious step was to integrate the top-performing model into a real-time expert decision support system after identifying TRF as the top performer. In the following, with the application of KF tool, an expert system is designed to predict the adsorption performance and process control as an alarm management system. Fig. 6(a) illustrates a comprehensive workflow for constructing and evaluating a machine learning model using a visual programming tool with WEKA. The process begins with the CSVLoader, which imports the dataset from a CSV file. This dataset is then passed to the ClassAssigner node, which designates the class attribute necessary for supervised learning tasks, setting the stage for the model training. Afterward, the Cross-ValidationFoldMaker node divides the dataset into training and testing sets through cross-validation. This method ensures that the model is validated on different subsets of the data, promoting a reliable estimation of its performance. The core of the modeling process is handled by the RandomForest node, which applies the TRF. This ensemble learning technique constructs multiple decision trees and aggregates their outputs to produce more accurate and stable predictions. To facilitate further evaluation and analysis, the PredictionAppender node attaches the predictions generated by the TRF model to the dataset. Following this, the RemoveWithValues node filters out instances with lower than 80 % removal efficiency of different heavy metal ions in the system. The model's performance is then rigorously evaluated by the ClassifierPerformanceEvaluator, which uses metrics like accuracy, precision, and recall to assess the classifier's effectiveness. All the analyses of the expert system are set on TRF with 80 % SP, which was optimized before. For visualization purposes, the ModelPerformanceChart node creates charts, such as ROC curves, to illustrate the model's performance graphically. Meanwhile, the ScatterPlotMatrix node generates a scatter plot matrix, enabling the visualization of relationships between different attributes within the dataset (Hall and Reutemann, 2025).

Fig. 6(b) is a scatter plot displaying the relationship between pH and mass (g) of adsorbent. The data points are color-coded according to their class values, as indicated by the gradient color bar at the bottom (RP), ranging from 33.76 % to 98.75 %. The points vary in size, suggesting different weights or frequencies. A significant concentration of data points is observed at higher pH values (around 4.5 to 5), with a wide range of masses. This shows that the data are clearly classified, with pH values of 4–5 corresponding to higher adsorbent masses, typically in the

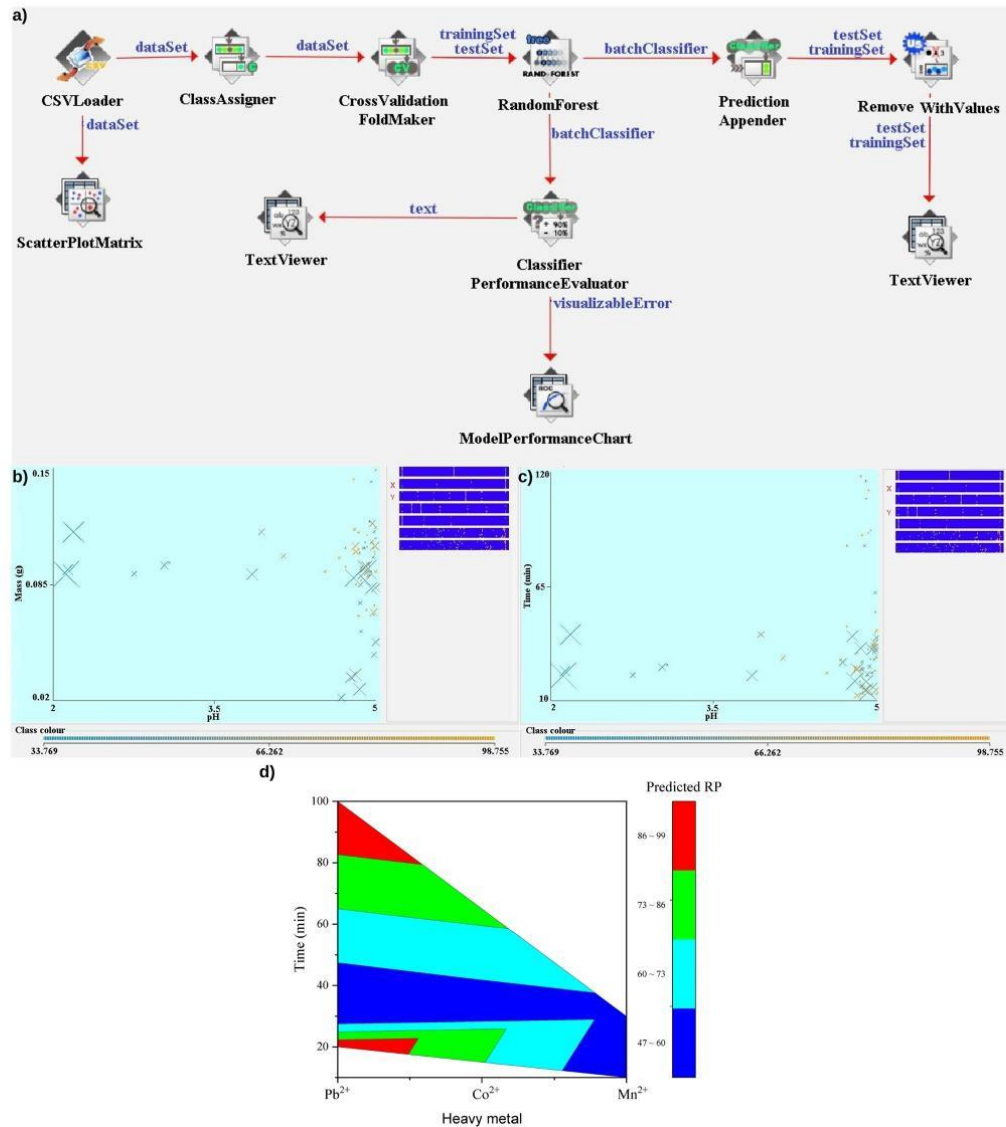


Fig. 6. The outputs of the designed expert system based on (a) schematic plan of the expert system, (b) classifications by TRF in pH vs. Mass of adsorbent, (c) classifications by TRF in pH vs. contact time, (d) control system analysis by time-heavy metal type.

0.10–0.15 g range, aligning with the finding that pH and mass are dominant factors controlling removal performance in LDC-based adsorption. Fig. 6(c) is another scatter plot showing the relationship between pH and time (min). Similar to Fig. 6(b), the data points are color-coded based on class values with the same color gradient. The distribution of points suggests that higher pH levels (around 4 to 5) correspond to a broader range of time values, from around 10 to 120 min. There are fewer data points at lower pH levels (around 2 to 3.5), indicating that these conditions are less common or less significant in the dataset. The largest RP values happen in the pH 4–5 range because

adsorption is chemically strongest there, where metal ions bond more successfully and the LDC surface is less protonated. In order to capture significant adsorption behavior, the majority of isothermal studies were carried out at pH 4–5. WEKA displays fewer data points in the pH 2–3.5 range since fewer experiments were conducted there. This diagram is the outcome of the model during the operation of the system, and operators can view this dashboard during water treatment as a monitoring tool to anticipate changes. The distribution highlights which pH and mass combinations are associated with higher or lower removal, enabling a better understanding of optimal conditions. The color gradient and

symbol size convey differences in removal performance, which is useful for refining process parameters. With the application of the KF concept, a dynamic model is presented to smarten the model. In fact, according to the unknown data in the operation concept, CSVLoader is connected to the application programming interface of telecommunication in the sensor network of water treatment plants, then the performance is predicted by the algorithm, and finally, according to undesirable performances of the heavy metal ions treatment, alarms will be informed to operators. For example, according to Fig. 6(d) is the output of the model. Considering Fig. 6(d), it can be found that the RP is categorized and the light and stronger blue colors are alarm zones for operators which they can be informed by some telecommunication protocols such as email or short message service (Ataei et al., 2024).

In the mentioned modeling, the experimental data is applied from experiments. The data is based on experiments, and in real water treatment processes, similar data could be provided in real time by sensors measuring parameters such as pH, IC, mass of adsorbent, and time. The RP of heavy metal adsorption can then be predicted using machine learning computations. The entire process can be implemented on processors such as Graphics Processing Units (GPUs), Field Programmable Gate Arrays (FPGAs), and others. Technologies in this field include Xylem's Decision Intelligence Platform (DIP) (Xylem US, 2025), Siemens' MindSphere IoT Platform for Water (Siemens Global, 2025), and ABB's Ability™ Smart Sensor Technology (ABB Group, 2025).

According to the earlier KF design and basic modeling workflow, the development of the expert system was further advanced in a programable environment to enable dynamic prediction and automated decision-making. Since the TRF method displayed greater accuracy and robustness during the initial MLA screening, the continuation of modeling was centered on TRF as the fundamental prediction engine. In the next phase, the adsorption performance values (RP) were categorized into four operating classes: [11 %–33 %], [33 %–55 %], [55 %–77 %], and [77 %–99 %], to enable multi-level categorization for process monitoring. Subsequently, key TRF hyperparameters (number of trees, minimum leaf size, and maximum splits) were tuned to optimize model performance. Finally, the dynamically optimized model was integrated into an alarm management system that flags operational risk conditions

using the threshold $RP < 80\%$, forming the basis of a real-time smart control module.

Fig. 7(a) showing the impact of the number of trees on predictive performance for the TRF model: with increasing number of trees (10–100), R^2 increases sharply and RMSE decreases, meaning better generalization; but after 100, both metrics start to slow down in changes indicating progressively smaller gains in accuracy and finally suggesting that circa hundred is a good compromise between accuracy and cost. The influence of the minimum leaf size on model performance shown in Fig. 7(b), the model is best in terms of R^2 and RMSE when a small leaf size (1–5) is used owing to the fact that the fine-grained decision regions can capture nonlinear adsorption regimes, and large leaf sizes result in fast performance degradation with low R^2 but high RMSE because of too simple tree structures, assigned because 1 is preferable for description of complex interplay governing LDC-mediated adsorptive behaviors. Similarly, Fig. 7(c) highlights the sensitivity of the TRF model to the number of maximum splits, where R^2 fluctuates across the evaluated range but reaches its peak near 75 splits with a corresponding minimum in RMSE; values significantly above or below this threshold compromise predictive stability through underfitting or overfitting, indicating that moderate tree depth is ideal for representing the combined effects of pH, metal type, initial concentration, adsorbent mass, and contact time.

Fig. 7(d) summarizes the model's classification performance over four RP categories after hyperparameter adjustment and for test results. The confusion matrix demonstrates great accuracy for the top classes [55 %–77 %] and [77 %–99 %], which dominate the dataset and correlate to consistent adsorption circumstances. Misclassifications primarily occur in the lower classes, where fewer samples and overlapping adsorption patterns impede separability. However, the model consistently detects operational states with high efficiency, which is essential for intelligent process monitoring. Model accuracy for continuous RP prediction is further proven in Fig. 7(e) for the training dataset. TRF successfully captures the nonlinear relationships regulating heavy metal adsorption, as evidenced by the close clustering of points around the perfect-fit line ($R^2 = 0.9242$). This demonstrates the superiority of TRF over linear models, which lack the flexibility to reflect

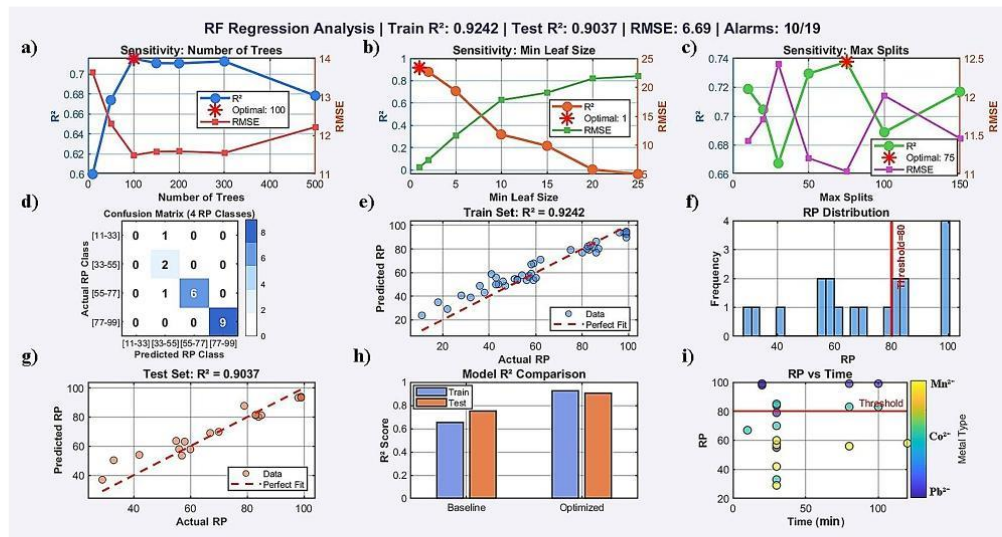


Fig. 7. TRF sensitivity and performance: (a) optimal tree count (=100); (b) best accuracy at leaf size = 1; (c) optimal splits = 75; (d) RP-class confusion matrix; (e) train R^2 ; (f) RP distribution with 80 % alarm threshold; (g) test R^2 ; (h) improved R^2 after tuning; (i) time-RP trends with alarm detection in low-performance.

multivariable interactions in CDW-derived adsorbents. Comparable performance is shown in the test dataset (Fig. 7g), where an R^2 of 0.9037 shows high generalization and low overfitting. The RMSE of around 6.69 is suitable for real-time environmental applications, where continual variations in input parameters are expected. In the following, The distribution of expected RP values in relation to the 80 % alert level is shown in Fig. 7(f) to facilitate real-time decision-making. The program properly recognizes a fraction of circumstances where RP falls below 80 %, delivering proactive alerts and relevant changes. This threshold-based approach is the foundation of the alert management module of the expert system.

The optimized TRF model outperforms the baseline configuration for both training and test sets in Fig. 7(h), which provides additional validity of the optimization technique. The diminishing gap between train and test R^2 values shows better generalization and reduced variance, illustrating the necessity of regular hyperparameter adjustment for preserving stability in real-time deployments. Lastly, a dynamic representation of RP performance over time for various metal ions is shown in Fig. 7(i). While Mn^{2+} shows more fluctuation and more frequent excursions into suboptimal regions, Pb^{2+} continuously stays above the 80 % threshold, indicating its significant affinity for LDC. These temporal patterns, together with metal-specific behavior, indicate the strength of the expert system in recording real-time aberrations and generating warnings when RP decreases below operational expectations. Collectively, Fig. 7(a-i) demonstrates that hyperparameter adjustment greatly boosts TRF prediction performance and stability, while the four-class RP categorization supports operational decision-making. The alarm subsystem accurately identifies instances where $RP < 80\%$, providing timely intelligence for intervention. Altogether, the integrated expert system indicates tremendous promise for real-time monitoring and automated control in Industry 4.0-oriented water treatment applications.

According to Fig. 8, for large-scale implementation of the presented smart control system, the soft-sensor system is built into a three-tier architecture consisting of the physical sensing layer, the edge-computing and data-integration layer (TRF model), and the cloud-based decision and control layer (alarm system). In the sensing layer, online instruments including pH probes (Uppuluri et al., 2023),

flow-based adsorbent-dosing controls (Besenhard et al., 2017; Dosing pump), reaction-time meters, and heavy metal concentration analyzers (Voda instrum; Zou et al., 2009), continuously send real-time data. These devices interact through conventional industrial protocols such as Modbus-Transmission Control Protocol (TCP) (Pricop et al., 2017), 4-20 mA analog signal (Dominguez-Nicolas et al., 2012)/Recommended Standard (RS)-485 (Lv and Wang, 2010), or Open Platform Communications-Unified Architecture (OPC-UA) (Vimos and Sacoto Cabrera, 2018), ensuring interoperability among varied treatment units. The captured signals are transmitted into the edge layer where a dedicated industrial gateway or microcontroller (e.g., advanced reduced instruction set computer machine: ARM-based unit) (Rahman et al., 2024) performs initial filtering, timestamp synchronization, and formatting.

These pre-processed data are subsequently transferred to the computational hardware either a local server or cloud platform, using secure Message Queuing Telemetry Transport/HyperText Transfer Protocol Secure (MQTT/HTTPS) protocols (Colitti et al., 2011; Hunkeler et al., 2008). The TRF-based soft-sensor accepts structured feature inputs (pH, adsorbent mass, heavy metal type and concentration, and contact time) and conducts continuous prediction of RP while simultaneously evaluating alert thresholds ($RP < 80\%$). When sub-optimal performance is observed, the system delivers control recommendations to the dosing pump and reactor controller, and simultaneously receives RSM-optimized values for pH and adsorbent mass for corrective action. All data streams, prediction outputs, and control actions are logged in a uniform dataset for periodic model retraining. This modular communication architecture facilitates the large-scale Industry 4.0 implementation of intelligent adsorption-based water treatment and enables smooth connection with current Supervisory Control and Data Acquisition (SCADA) (Ahmad Zaki Hamidi et al., 2018) systems.

The application of expert systems in the adsorption process as a DSS is rare (Krzywanski et al., 2024). While, in the following, some different DSS in water/wastewater processes is discussed.

The DSS in this study, utilizing WEKA/MATLAB for machine learning and the Knowledge Flow platform for decision-making, handles data through descriptive statistics, RSM for sensitivity analysis, and MLAs for predictions. It controls parameters such as pH, IC, adsorbent mass, and

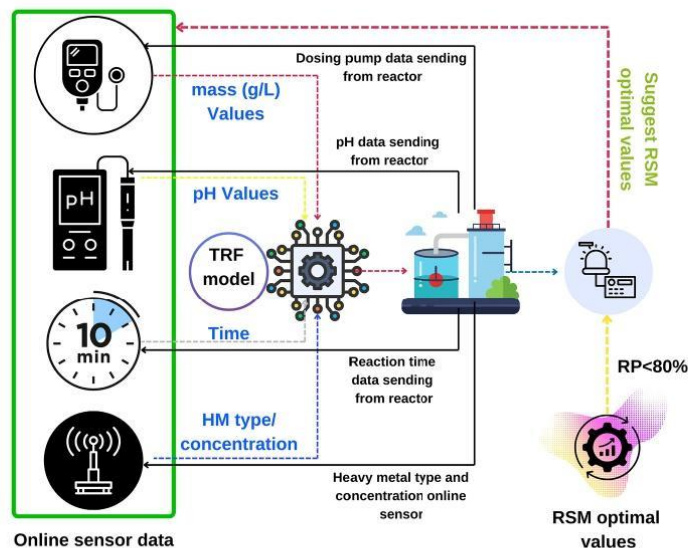


Fig. 8. Conceptual model of the smart control system for heavy metal adsorption based on the MLA-RSM framework developed in this study.

time, with removal percentages as outputs. In contrast, the expert system by Puñal et al. (2002) employs Visual Basic, Access, and MATLAB for software integration, focusing on real-time monitoring and fault diagnosis in anaerobic wastewater treatment, managing variables like gas flow, pH, and temperature to predict system states and provide operator recommendations.

In this study, the DSS uses descriptive statistics, RSM, and MLAs for predictions, controlling parameters like pH, IC, adsorbent mass, and time, with removal percentages as outputs. In contrast, Krzywanski et al. (2024)'s study develops an AI-based Diagnostic (AID) system using an AutoMLAs approach for adsorption cooling and desalination systems. They employed the DataRobot platform to select the best predictive model from 42 different models, focusing on 42 input parameters. The AID system achieved high efficiency, with an Area Under the Curve (AUC) of 0.988, RMSE of 0.2, and LogLoss of 0.14, and accurately predicted system health and failure states. While this study focuses on heavy metal adsorption with specific optimization, Krzywanski et al. (2024)'s work emphasizes predictive maintenance in cooling and desalination, showcasing broader applications and higher prediction accuracy. Hu et al. (2017) demonstrated a sensor-free electrothermal swing adsorption system using Activated Carbon Fiber Cloth (ACFC), controlled via electrical resistance changes. While innovative, the study relied on fundamental simulation rather than machine learning, limiting real-time implementation feasibility. Mursic et al. (1996) used a LABVIEW-based interface to precisely operate an automated adsorption isotherm equipment. Nevertheless, the system lacks adaptability for real-time decision support system applications and is based on basic simulation rather than machine learning.

4. Conclusion

Adsorption is considered a suitable method for high concentration wastewater treatment, especially for heavy metals, yet its efficiency and automation require further improvement for industrial applications. This study focuses on optimizing the parameters for removing Pb^{2+} , Mn^{2+} , and Co^{2+} from water using LDC waste as an adsorbent by applying RSM. Based on the experimental data and RSM optimization, pH was found to be the main factor, with the optimum occurring close to pH 5, which is the greatest practical value without precipitation danger, even though the overall best conditions depend on the combined impacts of all variables. For Pb^{2+} , pH 5, 0.13 g of adsorbent, 33.5 min of contact time, and a starting concentration of 35 mg L⁻¹ were the ideal conditions. The ideal parameters for Co^{2+} and Mn^{2+} were pH 5, 0.15 g of adsorbent, starting concentrations of 10 mg L⁻¹, and contact periods of 110 and 120 min, respectively. The optimal conditions for each metal within the examined operational range is defined by these combined parameters. Furthermore, adsorbent mass and contact time also influence removal efficiency, though their effects are metal-specific. In this stage of the study, it is shown that of the twenty different algorithms tested, TRF and FMLP, along with the lazy IBK, were the most accurate for predicting RP during the adsorption process. Adjusting these algorithms shows the TRF algorithm works best at an 80 % split percentage, as indicated by a CC of ~0.99. Finally, a DSS with KF concepts is developed to manage and control alarm settings for heavy metal contamination removal in water treatment facilities. The DSS based on the TRF model in WEKA's KF platform predicted Pb^{2+} removal efficiency and provided real-time alerts for RP values below 80 %. These alerts, using automated thresholds and data mining, were set for smart control, enabling automated operational control in smart water treatment systems. As well, using real-time data, the DSS was capable of predicting in a range of 30–100 % heavy metal decontamination efficiency.

To forecast adsorption capacity, model kinetics, and enable real-time decision-making, future research should employ advanced machine learning models with continuous sensor-based data. Techniques such as deep forest models, neural networks, and XGBoost can enhance screening, mechanism interpretation, and optimization. Additionally,

integrating metaheuristic approaches like NSGA-II with machine learning may further improve performance and reduce prediction errors. This study is limited by the inherent variability of waste-derived adsorbents, which affects consistency. Algorithm performance should also be validated in real water treatment plants with plug-flow conditions, and larger real-world datasets are needed to strengthen reliability and accuracy.

CRedit authorship contribution statement

Mohammad Gheibi: Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Project administration, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Seyyed Roohollah Masoomi:** Writing – original draft, Software, Data curation, Conceptualization. **Mohammad Eftekhari:** Writing – review & editing, Validation, Methodology, Formal analysis, Data curation, Conceptualization. **Martin Palusák:** Writing – review & editing, Validation, Conceptualization. **Daniele Silvestri:** Writing – review & editing, Methodology, Investigation, Conceptualization. **Miroslav Černík:** Writing – review & editing, Resources, Project administration, Funding acquisition. **Stanisław Wacławek:** Writing – review & editing, Validation, Supervision, Resources, Project administration, Methodology, Funding acquisition, Conceptualization.

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.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.dche.2025.100286.

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4.2.2. Decision-Support Framework for Intelligent Heavy Metal Adsorption onto LDC

This study presents an AI-driven expert system for automating heavy-metal adsorption using low-density concrete waste as a sustainable adsorbent. A hybrid Artificial Neural Network-Genetic Algorithm (ANN-GA)-Monte Carlo framework was developed to predict, optimize, and stabilize adsorption performance for Pb^{2+} , Co^{2+} , and Mn^{2+} under nonlinear and uncertain operating conditions. The ANN model, trained on 66 experimental datasets with pH, adsorbent mass, contact time, and initial concentration as inputs, achieved strong predictive accuracy ($R^2 = 0.908$, RMSE = 7.43%). Using the trained ANN as a surrogate, GA optimization identified metal-specific optimal operating conditions, achieving maximum removal efficiencies of 100% for Pb^{2+} , 87.8% for Co^{2+} , and 71.62% for Mn^{2+} . Sensitivity analysis showed metal type and pH as the dominant control variables, while contact time and initial concentration had minor influence, enabling simplified automated control logic. Monte Carlo simulations (2000 runs, $\pm 5\%$ perturbation) quantified operational robustness, revealing low variability for Pb^{2+} (CV = 2.13%) and Mn^{2+} (CV = 3.29%), while Co^{2+} showed higher uncertainty (CV = 5.39%). The integrated system enables real-time optimization, uncertainty-aware decision-making, and digital-twin-ready automation of adsorption processes, addressing the primary limitation of conventional adsorption technologies: lack of intelligent process control. The following section provides a concise summary of the key aspects of this part of the thesis. The present study effectively addresses and satisfies **Research Objective 4**, filling the identified gap in the existing literature.

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AI-BASED EXPERT SYSTEM DESIGN FOR OPTIMISATION OF HEAVY METAL ADSORPTION ON WASTE-DERIVED ADSORBENTS

Abstract: Heavy metal contamination in wastewater is a major global problem that requires effective, automated, and sustainable treatment systems capable of linking laboratory-scale adsorption processes to intelligent large-scale operation. This work introduces a novel Expert System (ES) that combines a Genetic Algorithm (GA), Artificial Neural Networks (ANN), and Monte Carlo (MC) analysis for the automated removal of heavy metals using adsorbents obtained from Low-Density Concrete waste. This research is significant because adsorption systems are complex to automate due to their nonlinear behaviour and sensitivity to fluctuating operating conditions. By facilitating intelligent process control and optimisation, the system overcomes the main drawback of adsorption technology, which is its lack of automation. Experimental datasets for Pb²⁺, Co²⁺ and Mn²⁺ removal were used to train the ANN, which predicts removal from inputs (pH, adsorbent mass, contact time, initial concentration). GA then used this trained ANN to optimise these same factors. According to the sensitivity analysis, the most important control variables in heavy metal adsorption were heavy metal type, pH, and adsorbent mass. The ANN model demonstrated strong predictive performance ($R^2 = 0.908$, RMSE = 7.43 %), enabling GA to optimise process conditions and achieve removal efficiencies of 100 % (Pb²⁺), 87.8 % (Co²⁺), and 71.62 % (Mn²⁺) under identified optimal operating parameters. Operational reliability measured by MC simulations showed Co²⁺ had a higher variability (CV: Coefficient of Variation = 5.39 %) than Mn²⁺ (CV = 3.29 %) and Pb²⁺ (CV = 2.13 %), which had minimal uncertainty. Using resources from the circular economy, the ES provides a framework for a digital twin of an Industry 4.0-compliant water treatment system that enables translation of lab-based procedures into practical applications.

Keywords: expert system, heavy metal adsorption, machine learning, construction and demolition waste, water treatment plant, process automation, Industry 4.0

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Abbreviations	Descriptions
<i>ANFIS</i>	Adaptive Neuro-Fuzzy Inference System
<i>ANN</i>	Artificial Neural Networks
<i>AVG</i>	Average
<i>BP</i>	Back Propagation
<i>CDF</i>	Cumulative Distribution Function
<i>CI</i>	Confidence Intervals
<i>CNN</i>	Convolutional Neural Network
<i>CT</i>	Contact Time
<i>CV</i>	Coefficient of Variation
<i>DSS</i>	Decision Support System
<i>ES</i>	Expert System
<i>GA</i>	Genetic Algorithm
<i>IC</i>	Initial Concentration
<i>IoT</i>	Internet of Things
<i>KPI</i>	Key Performance Indicators
<i>LDC</i>	Low-Density Concrete
<i>MA</i>	Mass of Adsorbent
<i>MAD</i>	Median Absolute Deviation
<i>MAE</i>	Mean Absolute Error
<i>MC</i>	Monte Carlo
<i>MI</i>	Mutual Information
<i>ML</i>	Machine Learning
<i>MSE</i>	Mean Squared Error
<i>NMI</i>	Normalised Mutual Information
<i>PCA</i>	Principal Component Analysis
<i>PDF</i>	Probability Density Function
R^2	Coefficient of Determination
<i>RF</i>	Random Forest
<i>RMSE</i>	Root Mean Square Error
<i>RP</i>	Removal Percentage
<i>RSM</i>	Response Surface Methodology
<i>SD</i>	Standard Deviation
<i>SHAP</i>	Shapley Additive Explanations
<i>SVM</i>	Support Vector Machines
<i>UN SDGs</i>	United Nations Sustainable Development Goals

Introduction

With continuous technological progress and societal development, rapid industrialisation and urban expansion have led to an increase in the release of hazardous substances, particularly heavy metals [1, 2]. The presence of these contaminants adversely affects the quality of soil and water, disrupts associated ecosystems, and poses significant

risks to human health [3]. Compared with membrane, chemical, electrochemical, biological [4, 5] and photocatalytic methods, adsorption [6] remains the most cost-effective, well-established, and environmentally friendly technique for heavy metal [7-9] removal in wastewater treatment. Adsorption provides operational simplicity and high removal efficiency even at low metal concentrations. In contrast, membrane and electrochemical systems, though offering greater automation, are generally more complex and costlier. The main limitation of adsorption techniques is their limited ability to be automated, which restricts their continuous and large-scale application. However, adsorption continues to be a practical and extensively used method for the long-term removal of heavy metals [10]. Machine Learning (*ML*) plays a crucial role in automating adsorption processes for heavy metal removal by enabling accurate prediction, optimisation, and control of operational parameters [11]. Reducing experimental effort, algorithms such as Artificial Neural Network (*ANN*) [12], Support Vector Machines (*SVM*) [13], and Random Forest (*RF*) [14] can simulate nonlinear interactions between variables, e.g., pH, adsorbent dose, and contact time. The integration of *ML* with sensors and control systems enhances real-time monitoring, facilitates adaptive decision-making, and automates adsorption-based wastewater treatment effectively.

The automation, design, and predictive modelling of adsorption processes for the removal of heavy metals have been greatly enhanced by recent developments in *ML*. In order to discover important factors that have an impact on adsorption, such as synthesis and ambient conditions, Liang et al. [15] applied *ML* to Schwertmannite, a sea urchin-like adsorbent, using an *RF* model trained on 814 data groups with 27 attributes. Leng et al. [16] optimised pyrolysis settings using only biomass composition data by using a hybrid *ML* framework that combines relationships between biomass, production, biochar, and application to predict biochar characteristics and adsorption behaviour. In order to improve soil remediation techniques, Wang et al. [17] used a variety of *ML* models, most notably XGBoost, to pinpoint important soil characteristics that affect heavy metal adsorption, such as pH, reactive oxides, and organic matter. In a similar vein, Yaseen and Alhalimi [18] combined ensemble *ML* techniques including XGBoost, LightGBM, and Adaboost to forecast the adsorption efficiency of biochar-based heavy metal, emphasising the crucial roles of pH and metal-to-biochar ratios.

The adsorption capacity of geopolymers was predicted by Han et al. [19] using an Extra Trees model, which ranked feature importance based on parameters including pH, dose, and curing time. In order to simulate biochar sorption efficiency, Hassan et al. [20] used a combination of linear, nonlinear, and deep learning methods, such as *ANN*, Convolutional Neural Network (*CNN*), and CatBoost. They also used Shapley Additive Explanations (*SHAP*) analysis for automated performance evaluation and feature interpretation. Tank et al. [21] created amine-modified mesoporous silica, applied 14 *ML* algorithms using *SHAP*-based feature dependency analysis, and created a stacking regressor to improve adsorption performance prediction.

To forecast the ion exchange resin adsorption capacity for heavy metals, Xu et al. [22] created a library of resin adsorption data by combining LightGBM modelling with attributes gained from molecular simulations. Wang et al. [23] used regression, *ML*, and deep learning models to study heavy metal adsorption onto microplastics. *RF* produced the best prediction and identified environmental factors as the main adsorption determinants. Overall, these studies show how *ML* may revolutionise adsorbent synthesis, feature selection, and system

automation, enabling the development of intelligent and flexible adsorption-based decontamination systems.

The intricacy of waste-derived materials, whose heterogeneous and mixed surfaces make adsorption modelling and automation difficult [24], has not been addressed in previous studies that have applied *ML* to a variety of adsorbents. By combining metaheuristic algorithms with *ML* models, an Expert System (*ES*) framework is created in this study to bridge this gap. This results in a novel Decision Support System (*DSS*) that allows for automated control, prediction, and intelligent optimisation of heavy metal adsorption using waste-based adsorbents.

According to the identified research gap, the present study aims to develop a comprehensive dataset from experimental adsorption processes of heavy metals using Low-Density Concrete (*LDC*) [25] as the adsorbent. An *ML* model based on the *ANN* is then constructed to predict the Adsorption Performance. Subsequently, *GA* optimisation is performed using the trained *ANN* to identify optimal operating conditions. The model's uncertainty is evaluated through Monte Carlo (*MC*) analysis, and finally, its performance is validated under a simulated water treatment plant with synthetic data in the range of experimental conditions to assess the overall efficiency of the developed *ES*.

Materials and methods

The research program starts with gathering experimental data on heavy metal adsorption utilising adsorbents produced from construction and demolition waste (*LDC*), as shown in Figure 1. Significant parameters are then found by statistical analysis. Adsorption performance is predicted by the *ML* stage (*ANN*), and then important variables are optimised by using the meta-heuristic *GA*. Model uncertainty is then assessed using *MC* analysis. Lastly, all phases are integrated for automated adsorption process prediction, optimisation, and control using the built *ES*.

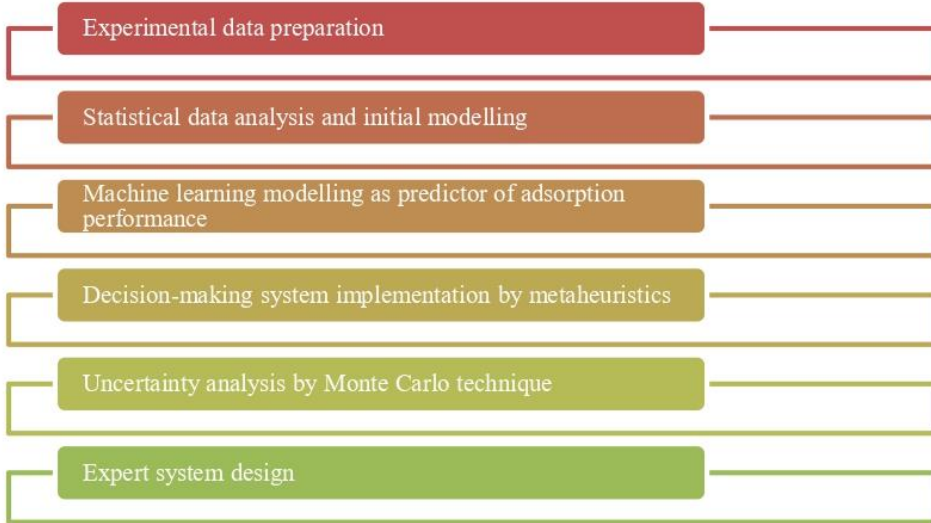


Fig. 1. The research roadmap of the present study

Experimental steps and dataset preparation

This study prepares the dataset based on the study of Gheibi et al. [25]. Experimental data on the adsorption of Pb^{2+} , Co^{2+} , and Mn^{2+} ions [26] (designated as 1-3) onto *LDC* adsorbent are included in the dataset. All experiments were conducted in single-contaminant reactors. Parameters including (1) pH (2-5), (2) Mass of Adsorbent (*MA*) (1.33-10) g/L, (3) Contact Time (*CT*) (1-120) min, (4) Initial Concentration (*IC*) of heavy metals (5-200) mg/L, and (5) Removal Percentage (*RP*) as the response are the five factors that control the system. One factor at a time, experiments were planned around a baseline (pH = 5, *MA* = 6.67 g/L, *CT* = 30 min, *IC* = 10 mg/L). Additionally, to assess the adsorption rate and equilibrium behaviour, kinetic analysis was performed by adjusting *CT* at constant *IC* = 5 mg/L and *MA* = 6.67 g/L. To ascertain the effects of the parameters, each metal system was evaluated in these controlled settings. The dataset consists of 66 adsorption test results, corresponding to 66 records with five input variables and one output variable. Consistency of dataset and nonlinear adsorption behaviour provides a strong basis for statistical modelling, *ML* prediction, kinetic evaluation, and optimisation using metaheuristic and uncertainty analyses in the *ES* that will be outlined below.

Statistical analysis

The statistical analysis in this study was methodically divided into four phases. The first step was data pre-processing for four parameters (pH, *MA*, *CT*, *IC*) as standardising input variables using z-score normalisation, detecting outliers using the Median Absolute Deviation (*MAD*) [27] approach. Second, the distribution of the data was summarised by computing descriptive statistical metrics as mean, standard deviation, quartiles, skewness, and kurtosis. Third, normalised Mutual Information (*MI*) [28] analysis was used to quantify inter-variable interdependence. Lastly, Principal Component Analysis (*PCA*) [29] was used to visualise patterns and reduce dimensionality. MATLAB R2019b was used to carry out all calculations and visualisations.

ANN-GA model structure

Figure 2 demonstrates the model structure for predicting the removal percentage (*RP*) of three heavy metal ions by *ANN* and maximising the *RPs* by *GA*.

Input parameters for creating the *ANN* include Metal type, pH, *MA*, *CT*, and *IC* to predict the *RP* of Pb^{2+} , Co^{2+} , and Mn^{2+} ions. Two hidden layers, each with 10 and 12 neurons, were used with the improved Levenberg-Marquardt training algorithm, or "trainlm" [30]. To guarantee strong generalisation, the dataset was split at random into 70 % for training, 15 % for validation, and 15 % for testing. The output neuron's activation function was set to linear, and the hidden layers' to tangent-sigmoid [30]. Iterative backpropagation was used to adjust the model parameters until the Mean Squared Error (*MSE*) was less than $1 \cdot 10^{-5}$. In this model, the cost function minimises the gap between *ANN*-predicted removal and the 100 % target, allowing *GA* to adjust input factors to maximise adsorption efficiency for each metal ion.

After developing the *ANN*, a *GA* [31] was applied to maximise *RP* by optimising the input variables within their physical ranges. The trained *ANN* was used as a surrogate model during the optimisation process. *GA* had a population of 100, a crossover rate of 0.8, 50 generations, and 10 elites. In this study, mutation was implemented using the default Gaussian mutation operator, which perturbs each decision variable by adding zero-mean

normally distributed noise, with variance controlled by the scale, shrink, and initial population range parameters to preserve diversity and avoid premature convergence [32]. The integrated *ANN-GA* framework [33] autonomously identified optimal operating conditions, ensuring low error and high predictive accuracy across all metal systems. Stochastic uncertainty quantification over the *ANN-GA* optimised adsorption response is carried out *via* the *MC* approach [34]. *MC* simulations (2000 iterations per ion) randomly perturb these ideal parameters within $\pm 5\%$ to provide probabilistic response distributions once the *ANN* has been trained and the *GA* has identified the best process conditions for each metal ion (Pb^{2+} , Co^{2+} , and Mn^{2+}). In the *MC* analysis, metal type, defined as a categorical variable (1 = Pb^{2+} , 2 = Co^{2+} , 3 = Mn^{2+}), is fixed within each *MC* run and is determined separately in an outer loop, not treated as a stochastic variable. Statistical descriptors, including mean (*AVG*), standard deviation (*SD*), Coefficient of Variation (*CV*), and (95-99) % Confidence Intervals (*CI*), are produced by recalculating the predicted *RP* using the trained *ANN* model in each iteration. By combining *MC*-driven uncertainty propagation, *GA*-based deterministic optimisation, and *ANN* predictive mapping, the framework allows for the assessment of model robustness and sensitivity to experimental modifications *via* visual dashboards such as *PDF* (Probability Density Function), *CDF* (Cumulative Distribution Function), and box plots. Note that all the declared computations were coded and run in MATLAB 2019b.

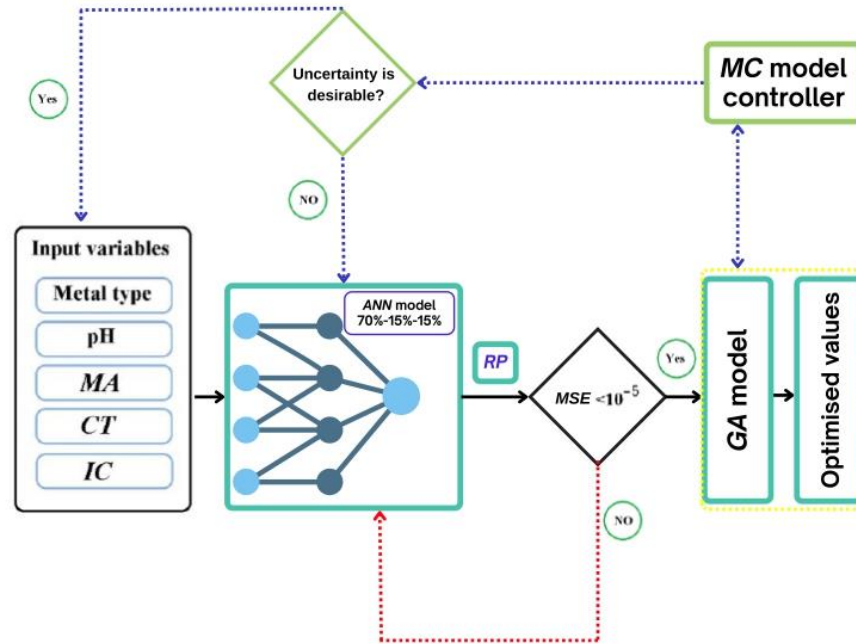


Fig. 2. The structure of *ANN-GA* model in the present study, Condition: *ANN*: Two hidden layers (10, 12 neurons); trainlm algorithm; tansig-hidden, linear-output; $MSE < 1 \cdot 10^{-5}$; split training: 70 % - testing: 15 % - validation: 15 % and *GA*: Pop 100; crossover 0.8; 50 generations; 10 elites; maximised *RP* $\rightarrow$ 100 % *via* optimised inputs

Expert system structure

An *ES* for intelligent control of metal ion removal in an industrial water treatment plant is formed by the implemented MATLAB R2019b software. To anticipate *RP*, it incorporates a feed-forward *ANN* trained on past process data, including Metal type, pH, *MA*, *CT*, and *IC*. To attain maximum *RP* for Pb^{2+} , Co^{2+} , and Mn^{2+} , these parameters are refined by *GA* optimisation. To simulate actual water treatment plant conditions, the system creates 200 synthetic sensor-based test datasets [35], allowing for online prediction and adaptive learning.

The synthetic online sensor dataset was generated within the experimentally validated operating domain by defining the input vector $x = [m, \text{pH}, MA, CT, IC]$, where the metal type $m \in \{1, 2, 3\}$ was treated as a fixed categorical variable and continuous parameters were sampled from bounded uniform distributions $\sim U(x_{i-\min}, x_{i-\max})$. Sensor uncertainty was emulated *via* multiplicative noise $x \sim i = x_i(1 + \epsilon_i)$, with $\epsilon_i \sim U(-0.02, 0.02)$ followed by range projection. Subsequently, *MC* analysis perturbed the *ANN-GA* optimised parameters within $\pm 5\%$ over multiple realisations to quantify process-level uncertainty around the optimal operating point. The 2 % simulated sensor uncertainty represents a controlled and reliable measurement regime, while the 5 % *MC* perturbation encompasses broader process and operational variability; thus, the smaller sensor noise is intentionally nested within the larger *MC* uncertainty to reflect the hierarchical nature of measurement and operational uncertainties.

The *ES* assesses operational dependability and model robustness using visual dashboards (response surfaces, Key Performance Indicators: *KPIs*, and process flow) and performance indicators (Coefficient of Determination: R^2 , Root Mean Square Error: *RMSE*, and Mean Absolute Error: *MAE*). While further work concentrates on *ES* performance validation under dynamic water treatment scenarios, the structure permits interfacing with sensors for autonomous optimisation.

Results and discussion

At the first stage, the specification of the dataset and available experimental results of Pb^{2+} , Co^{2+} , and Mn^{2+} adsorption onto the surface of *LDC* is analysed. A thorough summary of the features of the input dataset, inter-variable dependencies, and their overall variance structure relative to the *RP* is given in Figure 3.

The dataset was deemed complete, since there were no missing values in any of the response parameters (removal percentage) or input parameters (Metal type, pH, *MA*, *CT*, and *IC*). This validated the data's reliability for subsequent statistical analysis and modelling. Figure 3a shows the Normalised Mutual Information (*NMI*) matrix between the input variables. The off-diagonal *NMI* values remain below 0.12, indicating that each input delivers significantly independent information and confirming weak correlations among variables. A slight correlation (≈ 0.12) is observed between *CT* and *IC* in the adsorption test results. However, the categorical variable metal type exhibits negligible mutual information with all continuous variables, confirming minimal redundancy. Moreover, as metal type is a categorical variable, its low correlation with continuous parameters is expected and does not imply a lack of influence on removal efficiency. Therefore, the inclusion of all predictors in the modelling framework is justified, and the risk of multicollinearity is substantially reduced. Boxplots, which are a valuable tool for visualising central tendency, dispersion, and the existence of high-leverage observations, are used to display the input variable

distributions in Figure 3b. The pH variable has a clear left skew (skew = -2.81 , kurtosis = 9.9 , median = 5), with a few low extreme values. The *MA* variable exhibits a moderately left-skewed distribution (skew = -1.18 , kurtosis = 5.84), with most values clustered around 6.67 g/L and a limited number of high-leverage observations, indicating general consistency with occasional deviations. *CT* displays a broad and right-skewed distribution (skew = 2.08 , kurtosis = 6.41), spanning from 10 to 120 min, with 26 high-value influential observations. *IC* demonstrates the most pronounced non-Gaussian behaviour, with an extreme right skew (skew = 3.33 , kurtosis = 13.63), a dominant median of 10 mg/L, and nine high-leverage observations up to 200 mg/L, reflecting irregular concentration spikes.

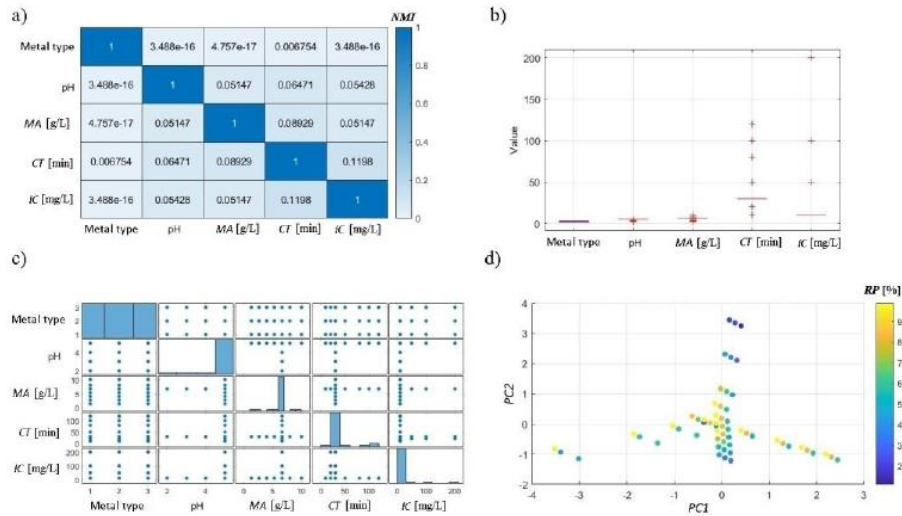


Fig. 3. Exploratory data analysis of input variables as per: a) normalised mutual information matrix according to weak inter-variable dependencies, b) boxplots as per skewed and heavy-tailed distributions with multiple high-leverage observations, c) Scatter matrix based on weak linear relationships among inputs, and d) *PCA* score plot ($PC1 = 23.8\%$, $PC2 = 22.1\%$) as per nonlinear variance patterns coloured by *RP*

The scatter matrix of input variables is shown in Figure 3c, enabling a visual examination of pairwise correlations. Consistent with the low *NMI* values (see Fig. 3a), the scatter plots exhibit diffuse and weakly structured patterns, confirming the absence of significant linear correlations. While *CT* and *IC* histograms along the diagonal indicate heavy-tailed distributions, Metal type and pH plots' vertical and horizontal bands are the result of their discrete or limited numeric levels. The absence of clear patterns indicates that potential correlations between these characteristics and *RP* are likely nonlinear and influenced by complex interactions rather than simple linear dependencies.

The first two Principal Components (*PC1* and *PC2*) account for 23.8% and 22.1% of the total variance, respectively, according to the *PCA* score plot displayed in Figure 3d. Together, *PC1* and *PC2* explain about 45.9% of the total variance, indicating that nearly half of the dataset variability is represented in the two-dimensional *PCA* space, while the

remaining variance is distributed across higher components. This further confirms the inherent complexity of the adsorption dataset. The combination of discrete (Metal type, pH) and continuous (MA , CT , IC) variables results in a cross-shaped pattern in the score distribution. It is implied that RP variation results from a number of interrelated elements rather than a dominant linear trend because the RP colour gradient does not correspond linearly with either component.

In general, Figure 3 shows that the dataset is statistically complete, with skewed and heavy-tailed distributions and low correlation among predictors. The nonlinear structure indicates that advanced methods such as ML and GA or their integrations are suitable for predicting RP and supporting automated optimisation of the adsorption process.

In accordance with Zhao et al. [36], who highlighted that heavy metal adsorption systems display multivariate yet weakly coupled nonlinearities prior to interaction effects predominating, enabling ML models to isolate governing parameters, the weak mutual information among input variables suggests minimal collinearity. Therefore, under these conditions, the application of ML models is essential for addressing this type of problem, as adopted in this study.

Figure 4 presents the $ANN-GA$ results, covering model diagnostics, optimisation outcomes, and prediction performance. With dispersion only rising at the highest removal levels, Figure 4a demonstrates a close parity between actual and predicted RP for all three metals ($R^2 = 0.908$, $RMSE = 7.43\%$, $MAE = 5.04\%$). As shown in Figure 4g, rapid error reduction occurs within approximately 5-8 epochs, and the stable test curve indicates minimal overfitting and good generalisation. Consistent with the heavy-tailed inputs previously noted, the residuals in Figure 4h exhibit a small mean bias (mean = 0.49), approximately homoscedastic variance, and are largely limited within $\pm 2\sigma$ ($\sigma = 7.47$), with a few pronounced deviations observed at high RP values ($> 50\%$).

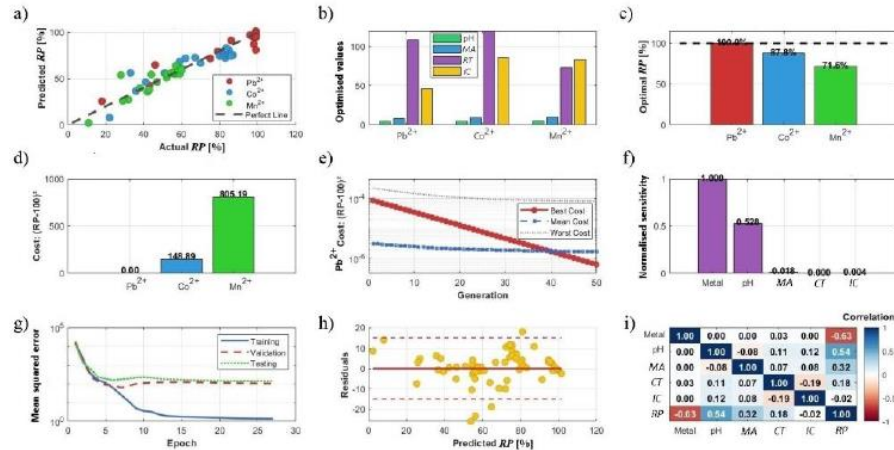


Fig. 4. $ANN-GA$ outcomes for ion removal based on: a) actual vs. predicted RP values, b) GA settings and optimal value suggestions for all input parameters (pH, MA , CT , and IC), c) maximum performance of RP per ion, d) cost function values per ion, e) convergence to best cost by 50 generations, f) sensitivity analysis of input variables, g) learning curves as per stable generalisation, h) residuals values, and i) Pearson correlation matrix of input/output parameters

The *GA* optimisation is reported in Figures 4b-e. Over 50 generations, the *GA* gradually reduces the best objective function (for instance, Pb^{2+} : Fig. 4e), keeping the best and mean costs apart and preventing protracted stalls, which shows ongoing exploitation with some population variety. In Figure 4b, high pH (near the top bound), *MA* $\approx$ (8.26-10) g/L, specific *IC* ($\approx$ 46.2-86.2 mg/L), and metal-specific *CT* (high for $\text{Pb}^{2+}/\text{Co}^{2+}$ and lower for Mn^{2+}) are the clustered optimal operating points. As shown in Figures 4c and 4d, Mn^{2+} reaches a maximum removal of 71.6 % and exhibits the highest optimisation cost function value (805.19), compared with Pb^{2+} , which achieves 100 % removal with zero cost, and Co^{2+} , which attains 87.8 % removal with a cost of 148.89.

These results indicate that Mn^{2+} requires process intensification or an expanded design space, as it is inherently more difficult to remove under the studied conditions. The results of the optimisation for critical parameters vs. *RP* are demonstrated in Table 1.

Table 1
The results of *ANN-GA* model optimisation based on experimental input data

Metal ion	<i>IC</i> [mg/L]	pH [-]	<i>MA</i> [g/L]	<i>CT</i> [min]	<i>RP</i> [%]
Pb^{2+}	46.2	4.8	8.26	109	100
Co^{2+}	86.2	5	9.99	120	87.8
Mn^{2+}	82.9	5	10	73	71.62

The interpretation of the model results in Figures 4f and 4i is the same. According to the normalised sensitivity analysis, metal type emerges as the dominant control variable (1.000), followed by pH (0.528) with a strong secondary influence, while *MA* (0.018), *CT* ($\approx$ 0), and *IC* (0.004) exhibit very limited direct sensitivity. Moreover, Figure 4i presents the Pearson correlation matrix, showing the relationships between *RP* and the other heavy metal adsorption variables. Most notably, *RP* exhibits a strong negative correlation with metal type (-0.63), indicating that as the metal type defined number increases (from Pb^{2+} to Mn^{2+}), *RP* values tend to decrease substantially. Conversely, *RP* demonstrates a moderate positive correlation with pH (0.54), suggesting that higher pH levels are associated with elevated *RP* values. Additionally, *RP* shows a weak to moderate positive relationship with *MA* (0.32), while *CT* displays only a minimal correlation (0.18). The *IC* parameter appears nearly independent of *RP*, with a correlation coefficient of -0.02 . Together, these diagnostics are used to support the *ANN* as a reliable surrogate model for *GA*-based search, and a practical strategy is suggested in which pH control is prioritised, *MA* is adjusted as a secondary lever, and *CT* and *IC* are treated as lower-order constraints.

The *ANN-GA* hybrid approach has demonstrated strong predictive capabilities across various adsorption systems. Fan et al. [37] achieved $R^2 = 0.99$ for Cd^{2+} removal using Reduced Graphene Oxide-Supported Nanoscale Zero-Valent Iron (nZVI/rGO) composites, while Halder et al. [38] reported $R^2 = 0.991$ for Cr^{6+} adsorption optimisation, and Mandal et al. [39] obtained $R^2 = 0.975$ for As^{3+} removal. The current study's $R^2 = 0.908$, while slightly lower, is attributable to the inherent variability of waste-derived adsorbents (*LDC*) compared to engineered materials, and single-metal batch experiments. Importantly, the current study extends beyond prediction accuracy to integrate *GA* optimisation and *MC* uncertainty quantification, providing operational robustness metrics (*CV* values) absent in prior *ANN*-only approaches. The identification of pH as dominant variable aligns with Mandal et al. [39] findings, confirming the reliability of the sensitivity analysis despite the moderate R^2 value.

GA convergence across metals is compared in Figure 5a-c. For Pb^{2+} (Fig. 5a), the best-cost value decreases rapidly from $\sim 10^{-4}$ to less than 10^{-6} ($\approx 99.3\%$ reduction in the cost function relative to the initial *GA* baseline) with a reduction of best-mean gap. Moreover, for Co^{2+} (Fig. 5b), the best-cost decreases nearly exponentially from $\sim \text{more than } 2 \cdot 10^3$ to 148.89 ($\approx 94.3\%$ improvement), indicating the stable convergence. Finally, for Mn^{2+} (Fig. 5c), the best cost remains high (805.19) despite a 47.5 % improvement, indicating a more difficult optimisation landscape.

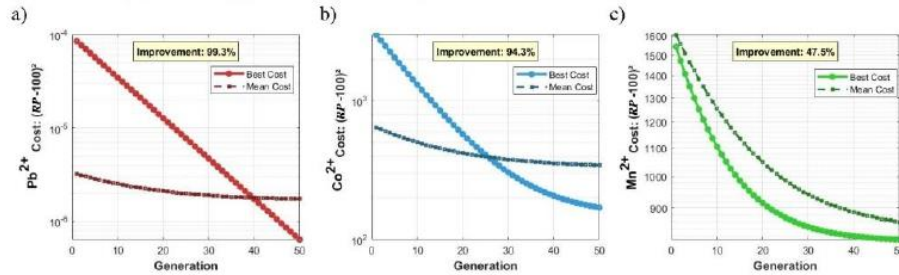


Fig. 5. *GA* convergence by metal: a) Pb^{2+} , b) Co^{2+} and c) Mn^{2+} . *GA* conditions: population size: 100, 50 generations, elite count: 10, crossover fraction: 0.80, fitness based on squared deviation of removal percentage from 100 %

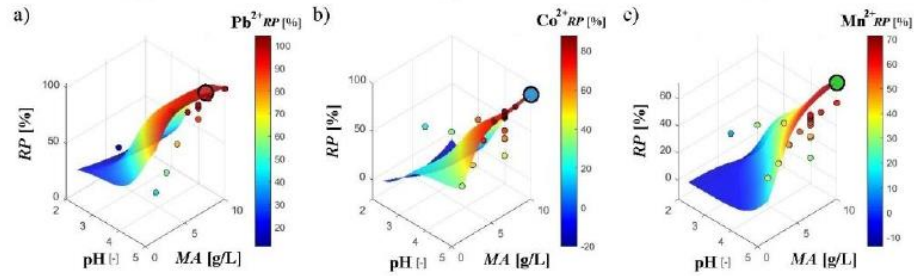


Fig. 6. Response surfaces (*RP* vs. *pH*, *MA*): a) Pb^{2+} (*CT*: 109 min, *IC*: 46.2 mg/L) and b) Co^{2+} (*CT*: 120 min, *IC*: 86.2 mg/L), and c) Mn^{2+} (*CT*: 73 min, *IC*: 82.9 mg/L) as per *ANN-GA* simulations, *GA* conditions: population size: 100, 50 generations, elite count: 10, crossover fraction: 0.80, fitness based on squared deviation of removal percentage from 100 %

For each Metal type at different specific *CT* and *IC*, Figure 6 plots the *ANN-GA* response surfaces of *RP* against *pH* and *MA*. As *MA* approaches approximately (6.67-10) g/L, *RP* increases markedly, with improvements near the lower acidity boundary ($\text{pH} \approx 5$) further enhanced by *pH* effects. At high *MA* and *pH*, Pb^{2+} shows near-complete removal across a broad region (Fig. 6a). Moreover, Co^{2+} exhibits a similar trend but with different *pH-MA* interaction effects (Fig. 6b). Besides, Mn^{2+} displays a smoother response and a lower removal plateau of about 70 % (Fig. 6c), indicating lower adsorption efficiency. Pb^{2+} has a lower hydration energy and acts as a softer Lewis acid, which favours strong inner-sphere complexation with alkaline *LDC* surface sites and permits incipient precipitation as $\text{Pb}(\text{OH})_2$ /basic carbonates at high *pH* [25]. This superiority is mechanistically feasible. These findings align with those of Macena et al. [40], who determined that variations in adsorption

affinity among heavy metals are affected mainly by ionic softness, hydration state, and the extent of complexation with surface functional groups.

The *ANN-GA* framework's model-based outputs for single factors vs. *RP*, are shown in Figures 7a-d. The effects of pH, *MA*, *CT*, and *IC* on *RP* collectively reveal a consistent adsorption hierarchy among the metal ions. Increasing pH enhances *RP* for Pb^{2+} and Co^{2+} , leading to high removal efficiencies at the upper pH range, whereas Mn^{2+} exhibits a weaker and more gradual response, with moderate removal efficiency at higher pH. In parallel, increasing *MA* strongly promotes *RP* for all metals, with Pb^{2+} and Co^{2+} showing a sharp rise, while Mn^{2+} requires higher *MA* values to achieve comparable performance. Prolonged *CT* has a limited additional effect, rapidly driving Pb^{2+} and Co^{2+} toward early saturation, whereas Mn^{2+} remains partially kinetically constrained. Conversely, increasing *IC* reduces *RP* due to active site saturation, with this inhibitory effect being most pronounced for Mn^{2+} because of its weaker surface affinity. From the standpoint of control systems, the main variables that should be changed (high loop gain) are pH and *MA*. Then, *CT* should be adjusted for each metal (longer for Co^{2+}). The outcomes illustrated in Figure 7d indicate that *IC* has the lowest influence on *RP* according to the *ANN-GA* model, exhibits an inverse relationship with *RP*, and, due to its fluctuations, should be specifically controlled for Co^{2+} at the optimal values reported in Table 1.

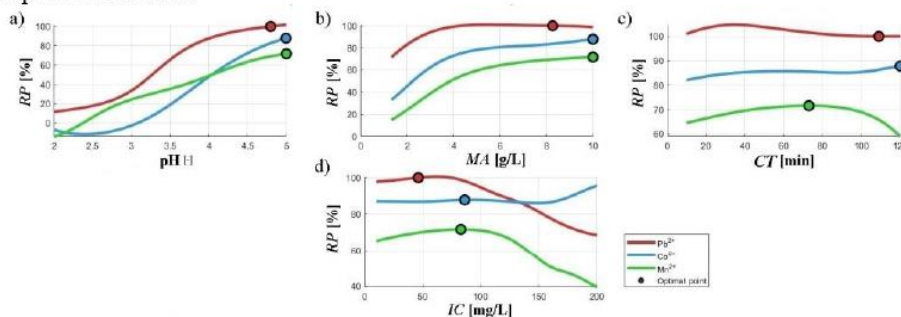


Fig. 7. *ANN-GA* response curves for Pb^{2+} , Co^{2+} , Mn^{2+} as per: a) pH vs. *RP*, b) *MA* vs. *RP*, c) *CT* vs. *RP*, d) *IC* vs. *RP*, *GA* conditions: population size: 100, 50 generations, elite count: 10, crossover fraction: 0.80, fitness based on squared deviation of removal percentage from 100 %. Note: In each plot, the remaining factors are fixed at their optimal values as listed in Table 1

Huang et al. [41] explained that precipitation, surface complexation, and ion exchange within the C–S–H and ettringite phases are necessary for heavy metal stabilisation in waste-based matrices, and the *GA* optimisation results following *ANN* prediction in this study are in good agreement with their findings. Co^{2+} and Mn^{2+} showed lesser fixation because they were present in more soluble forms, such as $\text{MnSO}_4 \cdot 2\text{H}_2\text{O}$, but Pb^{2+} demonstrated the highest stability through the development of insoluble hydroxide and phosphate precipitates ($\text{Pb}(\text{OH})_2$, $\text{Pb}_3(\text{PO}_4)_2$). Stronger adsorption and precipitation under alkaline conditions are confirmed by the observed pH-dependent enhancement in removal, which is in line with Huang's [41] solidification mechanisms.

Figure 8 quantifies the reliability of the optimal operating rules under input variability and complements the preceding *ANN-GA* results. *MC* uncertainty analysis of the *ANN-GA* model is reported in all panels.

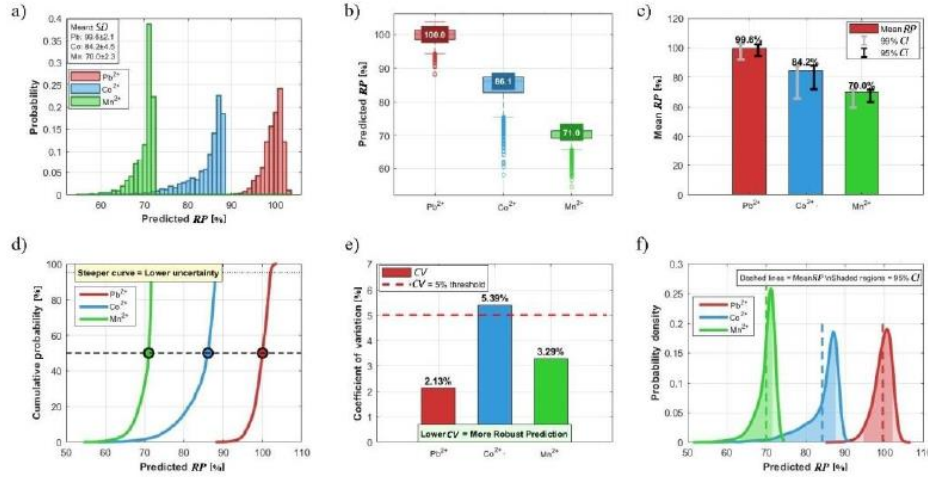


Fig. 8. ANN-GA-MC (2,000 runs) uncertainty: a) RP distributions, b) boxplots of MC analysis, c) means with 95/99 % Confidence Intervals (CIs), d) steep CDFs indicate robustness (Pb^{2+} , Co^{2+}), e) CVs in the three metals adsorption onto LDC, and f) PDFs to set control margins, MC Conditions: 2000 runs, $\pm 5\%$ perturbation of pH, MA , CT , IC , ANN-based RP uncertainty

After 2000 MC cycles at GA-recommended settings (Fig. 8a), uncertainty quantification reveals metal-specific variability patterns. By absolute standard deviation, Pb^{2+} (SD = 2.1 %) shows the lowest variability, followed by Mn^{2+} (SD = 2.3 %) and Co^{2+} (SD = 4.5 %). In the probability density histograms, Mn^{2+} exhibits the tallest peak due to its untruncated distribution around 70 %, while Pb^{2+} 's distribution near the physical 100 % limit appears broader due to edge effects, and Co^{2+} displays the widest spread centered at 84.2 %.

Boxplots of the MC-predicted RP values are shown in Figure 8b. Pb^{2+} confirms modest dispersion and consistent performance with a narrow interquartile range and few deviant values. Co^{2+} shows significant variability under uncertainty with a large interquartile range and several low-value outliers. Mn^{2+} exhibits a modest spread, much narrower than Co^{2+} but bigger than Pb^{2+} , indicating regulated but not insignificant variability. Further, Figure 8c compares the MC mean RP values with their corresponding 95 % CI. Pb^{2+} achieves a mean RP of 99.6 %, with a 95 % CI of [94.51 %, 102.31 %], confirming near-complete and highly reliable removal. Co^{2+} reaches a mean RP of 84.2 %, with a wider 95 % CI of [71.87 %, 87.94 %], indicating reduced robustness. Mn^{2+} exhibits a mean RP of 70.00 %, bounded by a narrower 95 % CI of [63.37 %, 71.62 %], reflecting moderate stability around its optimal value.

The CDFs of RP are shown in Figure 8d. The steep slope of the Pb^{2+} CDF indicates a high probability of achieving RP values close to the optimum with minimal uncertainty. Besides, the more gradual slope observed for Co^{2+} reflects greater variability and a higher likelihood of deviation from ideal performance. The Mn^{2+} CDF lies between these two extremes, indicating moderate robustness and resilience. Moreover, each metal ion's CV is shown in Figure 8e. The most reliable prediction is Pb^{2+} , which shows the least fluctuation (CV = 2.13 %). The moderately stable system under stochastic perturbations is Mn^{2+} , which exhibits moderate variability (CV = 3.29 %), but Co^{2+} surpasses the 5 % robustness barrier

($CV = 5.39\%$) based on the study of González-Fernández et al. [42]. The *PDFs* of *RP* are shown in Figure 8f, with shaded areas denoting the 95 % confidence intervals and dashed lines denoting the *MC* mean. Strong dependability and narrow control margins are confirmed by the crisp, high-density peak that Pb^{2+} displays. Because of its flatter and wider density profile, Co^{2+} needs more cautious operational management. Once more, Mn^{2+} exhibits intermediate behaviour, with reasonable uncertainty boundaries and a moderately concentrated density.

To assess parameter sensitivity in micellar-enhanced ultrafiltration, Lin et al.'s [43] *MC-ANN* model employed 1000 stochastic iterations. It achieved over 99 % metal rejection and identified pH and surfactant ratio as the dominating parameters with the least amount of model uncertainty. Similarly, to evaluate resilience and regulate variability, the current *ANN-GA-MC* paradigm combines evolutionary optimisation with probabilistic *MC* simulations.

The MATLAB-based system illustrates how *ML*, optimisation, and sensor-based analytics are integrated under the Industry 4.0 paradigm in the simulated operation of the *ES* within an industrial water treatment plant. The *ES* turns static adsorption modelling into a dynamic decision-support environment that can intelligently govern processes, as seen in Figure 9 as the online dashboard output. The average *RP* for Pb^{2+} , Co^{2+} , and Mn^{2+} ions is continuously forecasted on the real-time control dashboard, which is shown in the upper panel. Across 200 synthetic test scenarios with varied operational conditions, Pb^{2+} exhibits the highest average *RP* (69.4 %), followed by Co^{2+} (63 %) and Mn^{2+} (47 %), while *GA*-optimised conditions achieve maximum removals of 100 % (Pb^{2+}), 87.8 % (Co^{2+}), and 71.6 % (Mn^{2+}).

The simulation's varied operational variability is validated by the pH and sorbent mass distributions obtained from 200 synthetic sensor-based test datasets. Time and concentration are secondary under optimised regimes, while pH and sorbent mass dominate process sensitivity, as seen by the ideal parameter heatmap. The capacity of the system to generalise predictions under industrial noise and uncertainty is demonstrated by the *ANN* training and validation curves ($R^2 = 0.993$), which show consistent convergence with little overfitting. In this study, $R^2 = 0.908$ represents the *ANN* performance on training, validation, and testing, while $R^2 = 0.993$ is obtained within the *ES* using simulated sensor-based data (just training and validation). The observed difference ($\Delta R^2 \approx 0.085$) reflects the exclusion of unseen test data and the resulting evaluation under more stable, operator-controlled conditions. This difference does not imply a change in model structure, but rather the performance of the same *ANN* when assessed using training and validation data only.

From a smart operational perspective, the *ES* integrates predictive analytics with autonomous optimisation loops. The *GA* iteratively refines operational setpoints to maximise the *RP* of Pb^{2+} , Co^{2+} , and Mn^{2+} , with *ANN* outputs continuously integrated into the *GA*-based control logic. These results indicate that the system can adaptively regulate treatment parameters, maintaining process efficiency despite fluctuating influent quality.

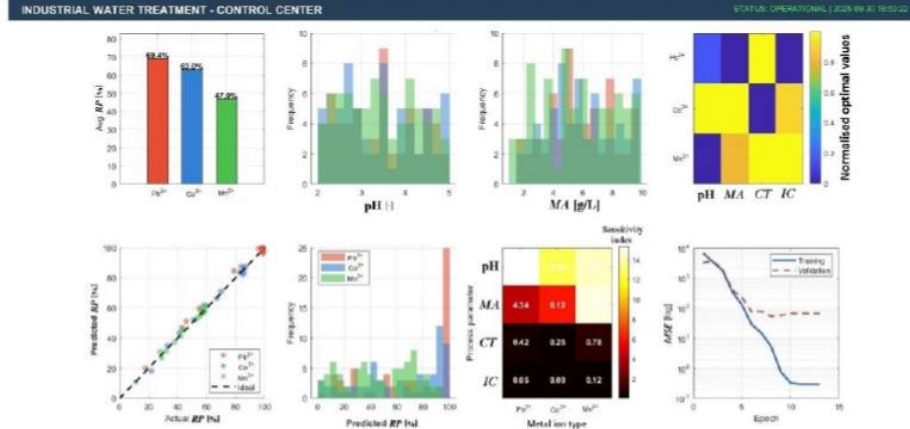


Fig. 9. Industrial water treatment control centre dashboard of the developed *ES* as per sensor-based data processing, *ANN-GA* predictive modelling, parameter optimisation, and intelligent monitoring within an Industry 4.0 operational framework

Figure 10 presents the online dashboard output, illustrating the optimisation outcomes of the *ES* through 3D response surface plots and contour maps for each metal ion. The surfaces visualise the nonlinear interplay between pH and MA on RP, illustrating that performance enhancement is primarily achieved within the high-pH, high-MA domain. The overlaid optimal points indicate the system's ability to autonomously discover the most efficient zones for operation.

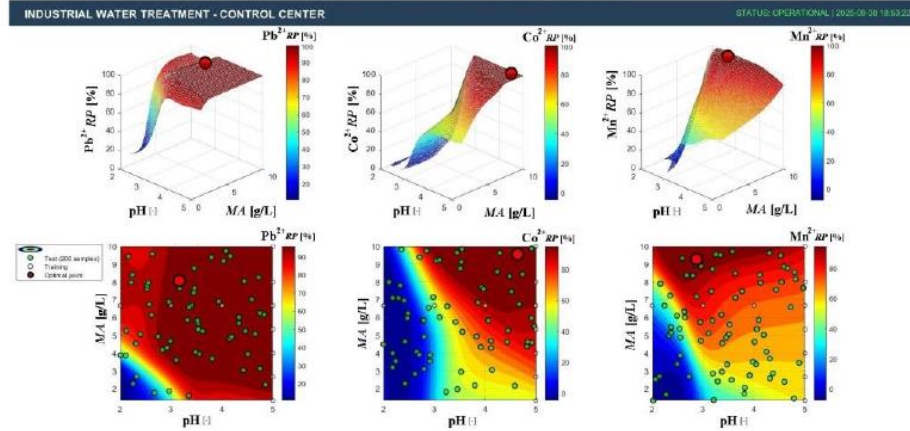


Fig. 10. Digital twin dashboard according to *ANN-GA*-based optimisation analysis, response surfaces, and optimal operational zones for Pb^{2+} , Co^{2+} , and Mn^{2+} adsorption in industrial level analysis

Using an *ANN*, *RF*, Radial Basis Function kernel, and Adaptive Neuro-Fuzzy Inference System (*ANFIS*), Pathania et al. [44] created an online *AI*-based wastewater monitoring

system that predicts pollutant adsorption in real time. However, their approach focused on prediction without optimisation. Wilson et al. [45] created a real-time biosorption monitoring system from an automation standpoint by combining an *ANN* model with an electronic tongue and a flow-injection potentiometric setup to assess transient signals from ion-selective sensors. This setup made it possible to identify several metals at once and partially automate the monitoring procedure. In order to facilitate optimisation and decision-making inside an Industry 4.0, the current study combines *ANN*, *GA*, and *MC* to advance automation.

By coupling *ANN-GA* predictive modelling with *MC*-based uncertainty analysis, the developed *ES* not only predicts and optimises removal efficiencies for Pb^{2+} , Co^{2+} , and Mn^{2+} but also quantifies operational robustness under realistic variations. The current approach changes adsorption modelling from a static optimisation or data-driven prediction tool into a decision-support system capable of real-time process control. This advancement aligns with the emerging concept of Virtual Laboratories [46], enabling digital experimentation, model validation, and process optimisation in simulated environments. Consequently, it establishes a scalable digital twin framework for sustainable water treatment systems. This could make it easier to connect with Internet of Things (*IoT*)-based sensing and real-time industrial automation in the future.

Conclusion

In this study, the *ANN-GA* framework predicted optimal removal efficiencies of 100 % for Pb^{2+} , 87.8 % for Co^{2+} , and 71.62 % for Mn^{2+} , with predictive accuracy of $R^2 = 0.908$ and $RMSE = 7.43$ %. *MC* analysis confirmed high operational robustness for Pb^{2+} ($CV = 2.13$ %) and Mn^{2+} ($CV = 3.29$ %), while Co^{2+} showed higher variability ($CV = 5.39$ %). By combining automation, uncertainty analysis, and process sustainability, the *ES* offers managerial decision assistance beyond technical optimisation, facilitating data-driven resource management and operational resilience in water treatment systems. By transforming construction and demolition waste into high-value functional adsorbents, the framework reduces reliance on virgin materials, reinforces circular resource utilisation, and contributes to UN SDGs 6 (Clean Water), 9 (Industry Innovation), and 12 (Responsible Consumption). In order to support sustainable process automation and informed policy development for environmental protection, the methodology converts static experimental insights into an eco-efficient digital twin framework. To improve sustainability, scalability, and circular economy integration in industrial water treatment, future research might expand the *ES* to dynamic multi-metal and real wastewater conditions, incorporate *IoT*-based real-time monitoring, and investigate hybrid organic waste-*LDC* composites.

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5. Conclusion

This thesis has provided a comprehensive and systematic investigation into the adsorption-based sustainable recovery of heavy metals from water resources using LDC derived from construction and demolition waste as a secondary raw material. By integrating mechanistic analysis, adsorption equilibrium and kinetic modeling, environmental impact assessment, and artificial intelligence-based automation, the study addresses critical scientific and technological limitations associated with conventional adsorption processes.

The first objective of this thesis was to elucidate, for the first time, the dominant physicochemical mechanisms governing the adsorption of Pb^{2+} , Co^{2+} , and Mn^{2+} onto LDC surfaces. The main findings demonstrate that:

- Heavy metal adsorption onto LDC is primarily governed by surface complexation reactions occurring on cementitious phases, particularly C–S–H and aluminosilicate structures rich in reactive hydroxyl groups.
- The dominant mechanism is inner-sphere complexation, where metal ions (Pb^{2+} , Co^{2+} , Mn^{2+}) form direct coordination bonds with deprotonated surface groups ($\equiv\text{Si}-\text{O}^-$ and $\equiv\text{Al}-\text{O}^-$), resulting in the formation of $\text{Si}-\text{O}-\text{M}$ and $\text{Al}-\text{O}-\text{M}$ linkages, as confirmed by FTIR peak shifts and the emergence of metal–oxygen vibration bands after adsorption.
- Outer-sphere electrostatic interactions contribute during the initial stage of adsorption, where hydrated metal ions are attracted to negatively charged surface sites without forming direct chemical bonds. This interaction is facilitated by the porous and heterogeneous structure of LDC, which provides accessible adsorption domains.
- A secondary mechanism involves cation exchange within the cementitious matrix, particularly substitution of Ca^{2+} and other native ions (e.g., Mg^{2+} , Al^{3+}) by incoming heavy metal ions. This is supported by elemental analysis showing a decrease in native cation concentrations after adsorption, indicating partial incorporation of metals into the solid structure.
- The adsorption process is further influenced by surface protonation–deprotonation equilibria, where increasing pH promotes deprotonation of surface hydroxyl groups, enhancing the availability of active binding sites and facilitating inner-sphere complexation, while low pH suppresses adsorption due to competition with H^+ ions.

The second objective focused on resolving the lack of comprehensive information regarding the isothermal and kinetic behavior of heavy metal adsorption on LDC. The results show that:

- Adsorption equilibrium for Pb^{2+} , Co^{2+} , and Mn^{2+} is best described by heterogeneous isotherm models, confirming non-uniform active site distribution on LDC surfaces.
- Kinetic analysis revealed that adsorption follows pseudo-second-order behavior, indicating that adsorption rate is controlled by surface interactions rather than simple mass transfer.
- The adsorption capacities of 43.1, 23.5, and 15.2 mg g^{-1} for Pb^{2+} , Mn^{2+} , and Co^{2+} , respectively, demonstrate that LDC performs competitively with many engineered adsorbents.
- The coupling of kinetic and equilibrium parameters with LDC porosity and mineral composition provides a robust framework for predicting adsorption performance under varying operating conditions.

The third objective addressed the insufficient integration of waste-derived adsorbents into environmental sustainability assessment frameworks. In this perspective:

- The application of the Sustainable Materials Management framework demonstrated that LDC-based adsorption offers clear environmental advantages due to waste valorization and avoided disposal impacts.
- Adsorption-normalized functional units enabled fair comparison between LDC and conventional adsorbents, highlighting the importance of performance-based sustainability metrics.
- Environmental hotspots were primarily associated with regeneration and auxiliary chemical use, emphasizing the need for optimized regeneration strategies.
- The results confirm that LDC-based adsorption aligns strongly with circular economy principles and sustainable water treatment objectives.

The fourth objective of this thesis was to overcome the limited automation and intelligence of traditional adsorption systems through the development of AI-based control strategies. The key contributions include:

- Machine learning analysis demonstrated that removal efficiency is governed by complex nonlinear interactions between operational variables, with models capturing dependencies that are not detectable through conventional statistical methods.
- Comparative evaluation of ML algorithms showed that tree-based and neural network models (TRF and MLP) exhibit superior generalization and predictive stability, indicating their robustness for modeling heterogeneous adsorption systems across different metal ions.
- Machine learning-based soft sensor models were developed and validated using experimental data, where TRF, lazy IBK, and Function Multilayer Perceptron achieved high predictive accuracy ($CC > 0.92$), significantly outperforming static RSM models for removal percentage estimation.
- This study developed an AI-based Decision Support System (DSS) that integrates ANN prediction, GA optimization, and Monte Carlo uncertainty analysis to support automated decision-making in heavy metal adsorption using LDC waste.
- The DSS employs a trained ANN as a surrogate process model to continuously predict removal efficiency from operational inputs (metal type, pH, adsorbent mass, contact time, and initial concentration), enabling GA-based identification of optimal operating setpoints under realistic constraints.
- By embedding Monte Carlo uncertainty propagation within the DSS, the system quantifies operational robustness and variability (via CV, confidence intervals, PDFs, and CDFs), allowing informed decisions on process stability, alarm thresholds, and reliability of optimized operating rules.

By addressing four clearly defined research gaps, this thesis contributes to a deeper scientific understanding of waste-based adsorption mechanisms, provides validated kinetic and equilibrium models for LDC, establishes a sustainability-oriented evaluation framework, and delivers an integrated intelligent control architecture for adsorption systems. Overall, the results confirm that LDC-derived adsorbents, when combined with advanced modeling and automation, represent a technically viable, environmentally sustainable, and digitally scalable solution for heavy metal removal from water and wastewater systems.

6. Outline of the future works

Future research focuses on sustainable E-waste management strategies that transform hazardous electronic residues into valuable resources within a circular economy framework. A promising direction involves the use of waste-derived adsorbents such as LDC for recovering metals like Ni^{2+} , Mn^{2+} , Zn^{2+} , and $\text{Fe}^{2+}/\text{Fe}^{3+}$ from bio-hydro metallurgically generated leachates. Biological or organic wastes digest E-waste materials to produce metal-rich leachates, enabling environmentally friendly recovery routes. Investigations emphasize the influence of parameters such as pH and adsorbent dosage on adsorption performance, while multi-model isothermal analyses; Langmuir, Freundlich, Temkin, and Dubinin–Radushkevich identify the governing mechanisms. Intelligent modeling approaches including Response Surface Methodology (RSM), ANNs, and optimization models like RSM provide predictive frameworks for adsorption capacity optimization under varying operational conditions. Further studies integrate environmental and sustainability assessments to evaluate the ecological and economic performance of waste-based adsorption systems. Environmental Impact Assessment (EIA) and Life Cycle Assessment (LCA) quantify resource efficiency, emission reduction, and overall environmental benefits compared to conventional techniques. Embedding these optimized materials into industrial E-waste treatment processes establishes a closed-loop framework where waste serves as a feedstock for environmental recovery, reinforcing the core principles of circular economy and sustainable resource utilization.

The next step of this thesis focuses on transforming the developed Expert System into a fully integrated Smart Decision-Support and Control Framework for autonomous wastewater treatment. Building on the GA-ANN-Monte Carlo platform, the future system will incorporate IoT-enabled monitoring, real-time data processing, and adaptive optimization to establish a digital twin that connects laboratory adsorption results with industrial operations. This next phase aims to scale the LDC waste-based adsorption system toward Industry 4.0 readiness, enabling intelligent automation, predictive control, and circular economy compliance for efficient, resilient, and sustainable heavy metal removal from water and wastewater.

7. Academic achievements

7.1. Publications

1. **Gheibi, M.**, Masoomi, S.R., Eftekhari, M., Akrami, M., Palušák, M., Silvestri, D., Černík, M. and Waclawek, S., 2025. Heavy metals adsorption using CDW adsorbents: A sustainable path for water purification. *Journal of Hazardous Materials Advances*, 20, p.100883.
2. **Gheibi, M.**, Masoomi, S.R., Eftekhari, M., Palušák, M., Silvestri, D., Černík, M. and Stanisław, W., 2025. Smart Control of Heavy Metal Adsorption onto LDC Wastes for Industry 4.0 Applications. *Digital Chemical Engineering*, p.100286.
3. **Gheibi, M.**, Hajiahmadi, Z., Jaromin-Glen, K., Eftekhari, M., Behzadian, K., Černík, M. and Waclawek, S., 2025. AI-based expert system design for optimisation of heavy metal adsorption on waste-derived adsorbents. *Chemistry-Didactics-Ecology-Metrology* 2025;30(1-2):47-65.
4. **Gheibi, M.**, Hajiahmadi, Z., Palušák, M., Silvestri, D., Černík, M., Eftekhari, M. and Waclawek, S., 2026. Turning E-waste into value: Innovative metal recovery using low-density concrete: Experiments and simulations. *Results in Chemistry*, p.103170.

5. **Gheibi, M.**, Palušák, M., Salava, M., Sydorenko, Y., Eftekhari, M., Silvestri, D., Černík, M. and Waclawek, S., 2026. A smart decision support system for Co²⁺ preconcentration using RSM and AI-Driven adsorptive modeling. *Engineering Science and Technology, an International Journal*, 77, p.102335.
6. **Gheibi, M.**, Waclawek, S., Leo, C.P., Sadr, S. and Behzadian, K., 2024, June. Dynamic Modelling, Simulation, and Sensitive Analysis of Lead Removal in a Fixed-Bed Adsorption Column using Waste-Based Materials. In *IOP Conference Series: Earth and Environmental Science* (Vol. 1368, No. 1, p. 012009). IOP Publishing.
7. **Gheibi, M.**, Masoomi, S.R., Magala, M.U., Fathollahi-Fard, A.M., Ghazikhani, A. and Behzadian, K., 2024. The Application of Artificial Intelligence (AI) in Adsorption Process of Heavy Metals: A Systematic Review. *Environmental Industry Letters*, 2(2), pp.57-78.
8. Enferadi, S., Eftekhari, M., **Gheibi, M.**, Moghaddam, N.N., Waclawek, S. and Behzadian, K., 2024. Modelling and optimising the performance of graphene oxide-Cu₂SnS₃-polyaniline nanocomposite as an adsorbent for mercury ion removal. *Environmental Science and Pollution Research*, 31(26), pp.38196-38216.
9. Makarem, A., Aldaghi, A., **Gheibi, M.**, Eftekhari, M. and Behzadian, K., 2024. One-step hydrothermal synthesis of a green NiCo-LDHs-rGO composite for the treatment of lead ion in aqueous solutions. *The Canadian Journal of Chemical Engineering*, 102(3), pp.1248-1261.
10. Hussien, L.B., Al-Kinani, E.M., Moradi, H., **Gheibi, M.** and Eftekhari, M., 2025. Folic acid capped Cu (II) nanocluster for solid phase extraction of lead from water and food samples. *Microchemical Journal*, 212, p.113374.
11. Enferadi, S., Moradi, M., Alkinani, A., Moradi, H., **Gheibi, M.** and Eftekhari, M., 2025. Synthesis of lanthanum phosphate–carbon nanoparticles using a hydrothermal method and their application for lead ion removal. *New Journal of Chemistry*, 49(12), pp.4864-4875.
12. Hussien, L.B., Al-Kinani, E.M., Hamud, W.M., Eftekhari, M., Moradi, H. and **Gheibi, M.**, 2026. Cu₂S-ZnS PANI nanocomposite adsorbent for the removal of lead ion from water samples. *Journal of Water Process Engineering*, 83, p.109501.

7.2. Projects

I actively contributed to the EU project “*Sustainable Remediation of Radionuclide Impacts on Land and Critical Materials Recovery*” with my research findings being integrated into the project’s overall framework and outcomes.

7.3. Conferences

1. **Gheibi, M.** et al. Intelligent Optimization of Heavy Metal Adsorption onto LDC Wastes [online]. Available from: <https://ecopole-conference.com/preliminary-programme/>, 34th Edition of the ECOpole Conference, 2025, Liberec, Czech Republic.
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3. **Gheibi, M.** et al. A novel perspective on heavy metal decontamination using waste-derived zeolite sludge composites [online]. Available from: <https://aquaconsoil.com/assets/250613-NWA-abstracts-presentaties-AquaConSoil-2025.pdf>.

4. **Gheibi, M.** et al. Prediction of Heavy Metal Emissions from Uranium Mines to Water Resources During Flood Disasters Using Fick's Second Law Modeling [online]. Available from: <https://meetingorganizer.copernicus.org/EGU25/EGU25-10555.html>.

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