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Comparative analysis of coagulants, turbidity-inducing materials, and Prussian blue removal efficiency for cesium

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Abstract. Cesium (Cs) contamination in water, particularly after nuclear incidents, poses significant environmental and health risks. This study aimed to evaluate and compare the effectiveness of various coagulants—alum, ferric chloride ($FeCl_3$), poly aluminum chloride (PAC), and poly aluminum hydroxide chloride silicate (PACl)—with and without turbidity-inducing materials, as well as the Cs removal efficiency of Prussian blue (PB). Laboratory-scale jar tests were conducted using raw water spiked with Cs-133 and analyzed using ICP-MS. $FeCl_3$ achieved the highest removal among coagulants (13.5%), while turbidity-inducing materials increased Cs removal to 37% due to increased particle-mediated adsorption and flocculation. PB demonstrated the highest efficiency (>99.9% removal at 10 mg/L), although its combination with PACl did not further enhance removal. These findings confirm PB as the most promising adsorbent for Cs removal and show that turbidity-enhancing strategies can improve coagulation outcomes. Further optimization of PB handling, immobilization, and large-scale implementation is needed to ensure safe and practical adoption in water treatment facilities.

Keywords: cesium; coagulation; prussian blue; radioactive pollution; turbidity

1. Introduction

South Korea's energy demand has steadily increased over the past two decades, with the country relying heavily on thermal power generated from imported coal, oil, and natural gas, which together account for over 80% of the energy mix [1]. Despite this reliance on imported fuels, energy consumption continues to grow, highlighting the country's low energy self-sufficiency. To address these challenges and reduce greenhouse gas emissions, South Korea is increasingly investing in nuclear and renewable energy sources. Nuclear power provides efficient electricity generation with relatively low carbon emissions; however, it also produces radioactive materials and waste that require careful handling to prevent environmental contamination and health risks [2]. This reliance on nuclear energy underscores the importance of developing effective strategies for managing radionuclides, such as cesium, in water and the environment.

South Korea's energy consumption has steadily increased over the past two decades. According to the '2022 Energy Statistics Yearbook' by the Ministry of Trade, Industry, and Energy, the country's per capita energy use rose from 4.20 TOE (ton of equivalent, 10^7 kcal) in 2001 to 5.94

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TOE in 2021 [3]. South Korea's primary energy sources include coal, oil, natural gas, hydropower, nuclear, and renewables. However, thermal power generation—relying heavily on coal, oil, and natural gas—makes up over 80% of the total energy mix. Despite this heavy dependence, South Korea imports 97% of the fuel required for thermal power generation, reflecting its low level of energy self-sufficiency [1]. A 2020 report by the International Energy Agency (IEA), 'Renewable Energy Information Analysis 2019, IEA/OECD,' noted that South Korea had the highest average annual growth rate in energy consumption among OECD countries but the lowest energy self-sufficiency [4].

Cesium (Cs) is a significant high-level component of aqueous radioactive waste streams, posing serious environmental and health risks due to its long half-life (approximately 30 years), high water solubility, and emission of gamma radiation [5]. Its resistance to degradation and tendency to bioaccumulate can cause physiological and genetic damage in living organisms, as observed in the aftermath of the Chernobyl (1986) and Fukushima (2011) nuclear disasters [6, 7]. Following the Fukushima nuclear accident in 2011, South Korea's Ministry of Environment introduced the 'Manual for Crisis Response to Nuclear Leakage Accidents in Adjacent Countries (Food and Water Sector),' which details methods for Cs removal from water treatment facilities [8]. As a result, there has been increasing interest in technologies aimed at eliminating Cs from water sources. The number of studies on Cs removal has grown significantly in recent years, highlighting the urgency of developing practical solutions for managing Cs contamination after nuclear incidents.

Currently, radionuclide-contaminated water can be treated through various physical, chemical, and biological methods [9]. These include adsorption [9, 10], extraction [11, 12], ion exchange [13, 14], chemical precipitation, and capacitive deionization [15]. Intensive chemical extraction techniques, such as ultrasonic-assisted leaching, have demonstrated high efficiency in recovering metals from concentrated sources [16], while sustainable vacuum distillation has been successfully applied for refining trace elements like selenium [17]. Similar selective separation strategies have been employed for recovering metals such as copper, tin, and lead from complex wastes [18], and particle interaction mechanisms have been shown to enhance removal efficiency in mineral processing applications [19]. Moreover, biological processes, including anaerobic microbial activity, can influence contaminant dynamics in environmental systems [20]. In regions like South Korea, where nuclear power constitutes a significant portion of the energy mix, waste heat from nuclear plants has potential for driving low-temperature desalination and water treatment systems, highlighting opportunities for integrated approaches to Cs removal [21].

Studies have identified coagulation and adsorption as effective techniques for removing Cs from water [22, 23]). The "Manual for Crisis Response to Nuclear Leakage Accidents in Adjacent Countries (Food and Water Sector)" highlights coagulation as a key method for Cs removal. To enhance this process in water treatment facilities, the manual recommends increasing coagulant dosages to 50–60 mg/L and adding 0.5–1.0 mg/L of chlorine to oxidize and disinfect radioactive substances. Additionally, injecting 10–30 mg/L of polyaluminum chloride (PAC) into raw water, with a contact time of at least 20 minutes, is advised. This method can achieve a maximum Cs removal efficiency of 46% through coagulation and precipitation, 94% with nanofiltration and reverse osmosis (RO), and up to 100% using ion exchange processes [24].

Laboratory experiments involving coagulation and precipitation were conducted using the Jar test with various coagulants, including alum, PAC, and high-basicity poly aluminum hydroxide chloride silicate (HIB-PAHCS). The results showed Cs removal efficiencies of less than 5% with alum and PAC, while HIB-PAHCS achieved over 20% removal from raw water with a turbidity of

50 NTU. Further improvements were observed by injecting 10 to 300 mg/L of zeolite, resulting in a maximum Cs removal efficiency of 91% [24, 25] In January 2016, the Seoul Water Research Institute evaluated Cs removal through coagulation, precipitation, and turbidity formation. The study found that coagulants alone were ineffective for Cs removal. However, Cs removal efficiency increased to 56% when turbidity reached 74 NTU [24].

Despite extensive research on Cs removal from groundwater, significant limitations remain in eliminating Cs solely through the addition of chemicals and additives without supplementary processes at existing water treatment facilities. This study aimed to evaluate the effectiveness of coagulants—alum, ferric chloride ($FeCl_3$), PAC, and poly aluminum hydroxide chloride silicate (PACl)—with and without the addition of turbidity-inducing materials. Additionally, Prussian blue (PB) adsorption was investigated as a highly effective method for Cs removal that does not require modifications to existing water treatment processes. Its application has the potential to influence tap water quality. The efficiency of these methods was verified through laboratory-scale experiments.

2. Experimental method

Cs removal experiments simulating water treatment processes were conducted in the laboratory using jar tests. The C-JT-H model from Changsin Science was used, with rapid stirring at 150 rpm for 5 min, followed by slow stirring at 50 rpm for 15 min. Samples were then allowed to settle for 30 min and filtered using a 47 mm glass microfiber filter (GF/C) prior to analysis. Cs concentrations were determined using an Inductively Coupled Plasma Mass Spectrometer (Agilent ICP-MS7500, USA) based on the Cs mass (m/z = 133) and calibrated using Li (m/z = 7, 6,340 cps), Y (m/z = 89, 12,554 cps), and Ti (m/z = 205, 7,698 cps).

Raw water was collected from the Nakdong River, with an average turbidity of 3.6–3.8 NTU and pH of 7.8–8.1. The natural Cs concentration in the river water was below 0.001 µg/L. For controlled experiments, the water was spiked to 100 µg/L using a Cs-133 standard solution (1,000 mg/L for ICP-MS). This elevated concentration, higher than typical real-world levels, was chosen to ensure accurate detection and reproducibility without affecting the fundamental removal mechanisms. Literature indicates that Cs concentrations following nuclear incidents are below 100 µg/L [26], supporting the relevance of this spiking level.

2.1 Coagulants and turbidity-inducing materials

Various coagulants, including alum, $FeCl_3$, PAC, and PACl were used in the experiment. These coagulants were prepared at concentrations of 5, 10, 20, 30, 40, 50, and 60 mg/L based on the Al_2O_3 for the Jar test. The coagulant dosage range of 5–60 mg/L was selected based on previous studies on cesium removal [24, 25] and practical guidelines outlined in the *Manual for Crisis Response to Nuclear Leakage Accidents in Adjacent Countries (Food and Water Sector)*, which recommends higher doses (50–60 mg/L) for enhanced coagulation. Preliminary jar tests conducted in this study also confirmed that dosages above 60 mg/L did not yield additional improvement, therefore 5–60 mg/L was chosen as the representative range. To assess the coagulation efficiency based on turbidity, turbidity-inducing materials such as Granite sand, Illite, and Zeolite with particle sizes of 200 mesh were used. The composition of each turbidity-inducing material is summarized in Table 1.

Table 1. Composition components of turbidity-inducing materials

	Granite sand	Illite	Zeolite
SiO_2	57.21	54.53	60.47
Al_2O_3	31.46	32.63	9.72
FeO_3	7.53	2.3	21.51
CaO	1.2	0.09	3.27
Na_2O	0.15	0.43	1.32
K_2O	0.98	8.65	2.85
P_2O_5	0.02	0.02	0.15
MgO	0.98	0.53	0.71
MnO	0.01	0.06	-
TiO_2	0.46	0.76	-

2.2 Composition of PB adsorbents

In the PB adsorption experiments, a $FeCl_3$ and potassium ferrocyanide mixture was used to synthesize PB by mixing at a ratio of 1:1. The efficiency of Cs removal was compared and analyzed using both insoluble and soluble PB. In this study, insoluble Prussian blue (PB) was used, which ensures stability in aqueous solutions and minimizes the risk of Cs desorption. Soluble PB was not employed because it could lead to residual PB in treated water and potential secondary contamination. Additionally, experiments were conducted to explore methods for increasing the removal efficiency when PB was used, such as examining the extent of Cs removal based on the contact time with PB.

3. Results and discussion

3.1 Effect of coagulant type and dosage

The Jar test was conducted by injecting the coagulants Alum, $FeCl_3$, PAC, and PACl at concentrations of 0, 5, 10, 20, 30, 40, and 60 mg/L, respectively, into raw water containing 100 µg/L of Cs. The results showed that $FeCl_3$ has the highest Cs removal efficiency among the other coagulants, achieving a maximum of 13.5% at 60 mg/L, while others exhibited lower, with results remaining below 10% regardless of dosage. The results showed that $FeCl_3$ has the highest Cs removal efficiency among the other coagulants, achieving a maximum of 13.5% at 60 mg/L, while others exhibited lower, with results remaining below 10% regardless of dosage. This enhanced performance of $FeCl_3$ may be attributed to the hydrolysis of Fe^{3+} ions, which generate positively charged hydroxide species that neutralize negatively charged colloids and promote floc formation. The freshly formed $Fe(OH)_3$ precipitates also present a high specific surface area, facilitating Cs^+ adsorption through electrostatic interactions and surface complexation. Although overall removal efficiencies were modest, these mechanisms likely underpin the relatively superior performance of $FeCl_3$ compared to alum, PAC, or PACl. Future work is needed to elucidate the speciation of Fe and its interaction with Cs in aqueous matrices to clarify the dominant removal pathways. The $FeCl_3$ achieves higher Cs removal than Al-based coagulants due to its higher charge density, more

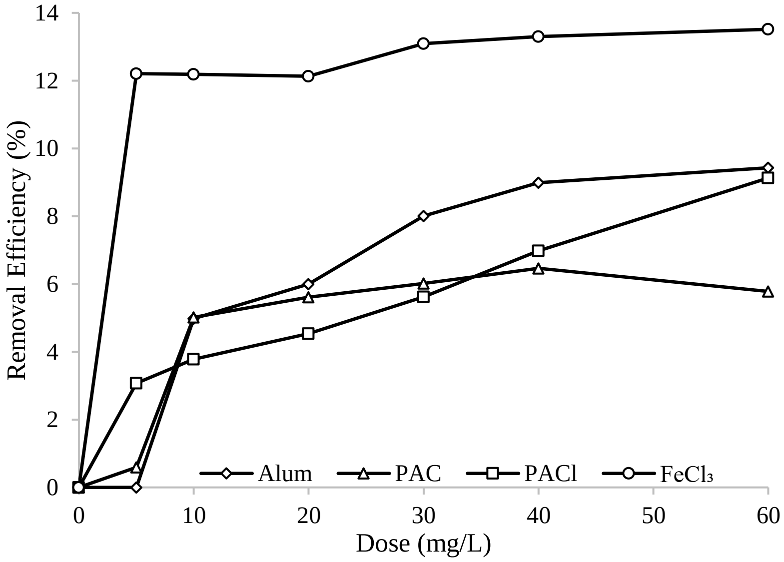

Figure 1. Effect of different coagulant dosages on Cs removal

effective hydrolysis products, and stronger formation of flocs that can adsorb Cs-laden particles.

These results suggest that increasing the coagulant dosage to approximately 50–60 mg/L slightly improves Cs removal efficiency. However, the overall removal remains modest, indicating that higher dosages do not substantially enhance performance. In addition, increasing coagulant dosage can influence the turbidity of raw water, potentially affecting downstream processes such as sedimentation or further treatment. Combined with the mechanistic insight that $FeCl_3$ hydrolysis and floc formation contribute to Cs removal via charge neutralization and surface adsorption, these findings highlight the need for careful optimization of coagulant type, dosage, and operational conditions to maximize treatment efficiency.

3.2 Effect of turbidity-inducing materials

Raw water from natural sources often exhibits turbidity due to soil and sediment contamination. Adding turbidity-inducing materials to simulate realistic conditions, offering insights for optimizing water treatment processes. Moreover, the literature suggests that increased turbidity enhances Cs removal [24]. Therefore, investigating the effects of turbidity-inducing materials can clarify their role in improving Cs removal efficiency under conditions imitating real-world scenarios, aiding in the advancement of water treatment methods. To examine the effect of turbidity, granite sand, illite, and zeolite were injected as turbidity-inducing material into raw water at concentrations of 10, 30, 60, 100, 150, and 200 mg/L. After adding turbidity-inducing material, the turbidity of water increased as the dosage of the turbidity-inducing material increased. For example, the initial turbidity of the raw water was 3.6 NTU, which increased to 7.2 NTU upon dosing with 10 mg/L granite sand and reached 120.4 NTU at 200 mg/L, with illite and zeolite showed changes in turbidity similar to those of granite sand.

Jar tests were conducted to investigate Cs removal using turbidity-inducing material, similar to the coagulation and precipitation experiments conducted using coagulants. After the jar test, the

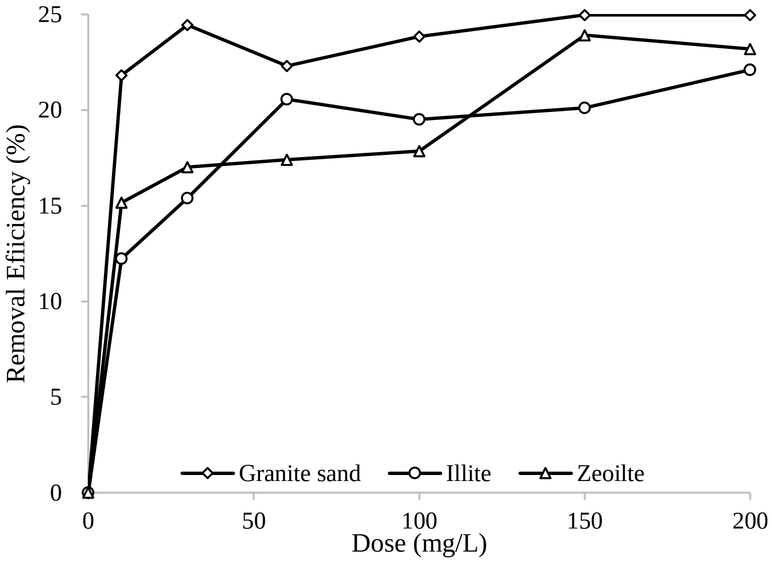

Figure 2. Analysis of Cs removal using granite sand, Illite, and zeolite

supernatant was collected, filtered through a GF/C, and analyzed using ICP-MS. The results are shown in Fig. 2, showing that when adding granite sand dosing at 10 mg/L into raw water in 100 µg/L of Cs, the removal was 21.83% and reached a maximum of 24.97% at 200 mg/L, with no significant change at dosing was higher than 10 mg/L. The zeolite's superior performance is primarily due to its intrinsic cation exchange capacity, which actively captures Cs ions, whereas granite sand and illite mainly contribute to turbidity and floc formation.

This indicates that the removal efficiency did not significantly improve as the concentration of granite sand increased further. The Cs removal using illite showed a similar pattern to granite sand, with a lower removal efficiency of 13.95% at 10 mg/L and a maximum of 17.99% at 200 mg/L. In contrast, zeolite was observed that Cs removal increased with dosage, with an efficiency range of 15.16 to 23.20% at 150 mg/L. However, no noticeable improvement in removal was observed at doses above 150 mg/L. The different behavior of zeolite is attributed to its unique properties in adsorption and ion exchange processes, which materials like granite sand and illite lack. This enables zeolite to remove Cs at lower dosages effectively. The Cs removal efficiency varies substantially among sorbents due to differences in ion-exchange and adsorption behaviors. Zeolitic materials, which possess high cation exchange capacity, often show enhanced uptake of Cs compared to common clays and inert sands due to their three-dimensional aluminosilicate framework and abundant exchange sites [27, 28]. Experimental comparisons of clay minerals indicate that illite and other clays adsorb Cs much less efficiently under environmental conditions than more selective sorbents [29]. These findings highlight the variability in performance among different turbidity-inducing materials and underscore the importance of optimizing their dosages for effective water treatment.

3.3 Effect of coagulants in turbidity-enhanced water

The experiment was conducted using the Jar test method, similar to the coagulant and turbidity

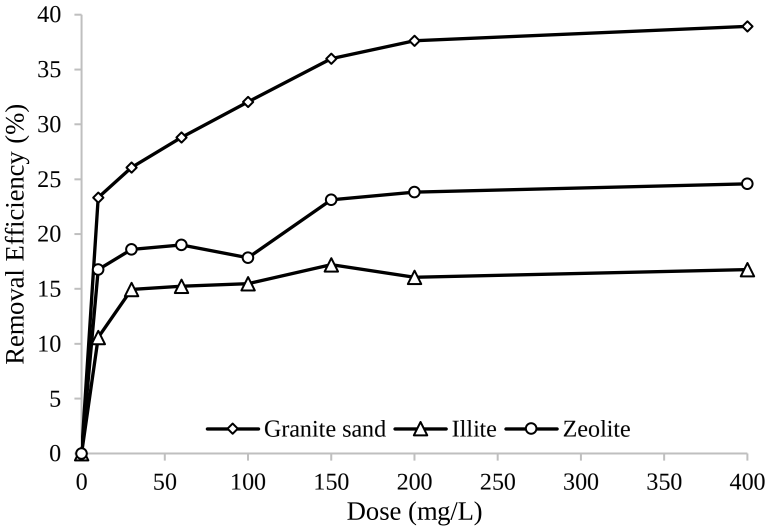

Figure 3. Analysis of Cs removal by granite sand, illite, and zeolite

induction experiment, where raw water containing 100 µg/L of Cs was subjected to turbulence for 10 min at 50 rpm after injecting the turbidity-inducing agent, with different dosages at 10, 30, 60, 100, 150, 200, and 400 mg/L and followed by the addition of PACl at 20 mg/L. The results showed that after inducing granite sand at 10 mg/L and adding PACl, Cs removal efficiency was 24%, which increased to 37% when the granite sand injection was 200 mg/L, as shown in Fig. 3. Indicated that when the coagulant was injected into the turbidity-formed raw water, the Cs removal increased as the turbidity of the raw water. This is because increased turbidity introduces more suspended particles, serving as bases for coagulant aggregation, forming larger and denser flocs. These flocs effectively trap Cs, enhancing removal efficiency during sedimentation. Additionally, the increased particles provide a greater surface area for Cs adsorption, further supported by coagulants that bind Cs-laden particles, facilitating their separation and improving the overall Cs removal process [24, 30]. On the other hand, Cs removal when using illite indicated 11.57% at 10 mg/L. It marginally increased to 13.57% at 200 mg/L, showing no significant difference observed in removing Cs after inducing turbidity using illite compared to the previous analysis, which only illite injection. Moreover, zeolite showed results similar to illite, which suggested that they may adsorb Cs before combining them with a coagulant, highlighting their selective adsorption properties. In conclusion, Cs removal increased with coagulant injection in raw water, where granite sand induced turbidity, but it did not improve when using illite and zeolite. This difference can be attributed to the compositions of these minerals. Illite and zeolite consist of specific minerals with consistent structures, and granite sand consists of various minerals with diverse structures, resulting in different ion adsorption capacities.

3.4 Cs removal using PB

PB is widely recognized for its high efficiency in removing Cs from water through adsorption. In this study, PB was prepared by mixing $FeCl_3$ and potassium ferrocyanide at a ratio of 1:1, and the effect of PB dosage was tested at concentrations of 1, 5, 10, and 20 mg/L. As shown in Fig. 4,

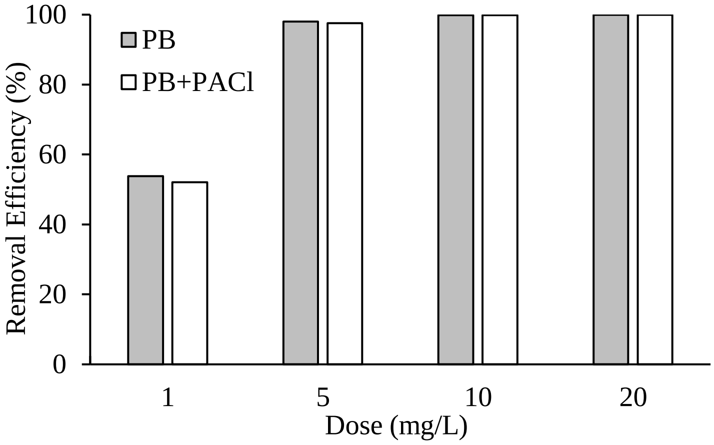

Figure 4. Effect of PB and PACl (@20 mg/L) on Cs removal

Cs removal efficiency increased from 53.8% at 1 mg/L to over 98% at 5 mg/L and reached a maximum of >99.9% at 10 mg/L, with no significant change at higher concentrations, indicating a saturation point [31]. To explore potential integration with existing water treatment processes, PB was combined with 20 mg/L PACl. The Cs removal efficiency did not significantly improve compared to PB alone. The presence of PACl may facilitate PB aggregation and floc formation, enhancing separation from treated water and reducing residual PB release. However, such aggregation may also reduce the available surface area for Cs adsorption, explaining the lack of improvement in removal efficiency. Future studies should investigate the balance between adsorption capacity and solid–liquid separation in PB-coagulant systems.

Although PB alone achieves near-complete Cs removal, environmental feasibility must be considered. PB is generally stable under neutral pH, minimizing Cs release. Nevertheless, residual PB particles may remain suspended, posing a risk of secondary contamination. Combining PB with coagulants or employing sedimentation/filtration can improve particle removal. Further studies on long-term stability, disposal strategies, and ecological impacts are necessary for safe large-scale application. Overall, these results indicate that PB alone is sufficient for optimal Cs removal. Using coagulants with PB does not enhance removal efficiency but facilitates easier separation, which is beneficial for minimizing secondary contamination and simplifying post-treatment processes, making this approach practical for large-scale water treatment applications.

For practical application, the recovery and immobilization of PB are important considerations. Techniques such as immobilizing PB on solid supports (e.g., silica, zeolite, or polymer matrices) have been reported to enhance recovery efficiency and minimize the risk of secondary contamination [32, 33]. In addition, while the integration of coagulants facilitates PB separation, it may also lead to sludge generation. Sludge management and safe disposal represent a scalability challenge for large-scale applications, particularly under emergency scenarios involving nuclear accidents. Therefore, future work should focus on evaluating PB immobilization strategies, sludge handling, and the overall feasibility of implementing PB-based systems in water treatment facilities.

While this study provides a comparative evaluation of coagulants, turbidity-inducing materials, and Prussian blue (PB) for Cs removal, several limitations exist. First, laboratory-scale jar tests may not fully replicate the dynamics of full-scale water treatment plants, and scaling up could introduce hydrodynamic and mixing challenges. Second, the study used spiked water with Cs-133 at concentrations higher than those typically observed in environmental contamination to ensure accurate detection, which may not reflect actual field conditions. Third, while PB demonstrated excellent removal efficiency, residual PB particles and long-term stability in diverse water chemistries remain uncertain. Finally, coagulant-turbidity interactions may vary depending on water quality parameters such as pH, organic matter content, and ionic strength, introducing variability in removal efficiency. These factors highlight the need for pilot-scale studies and further investigation to validate the practical applicability and safety of the proposed methods.

4. Conclusions

This study evaluated the effectiveness of conventional coagulants, turbidity-inducing materials, and Prussian blue (PB) for cesium (Cs) removal from water. Among the coagulants tested, $FeCl_3$ achieved the highest Cs removal efficiency (13.5%), while alum, PAC, and PACl were less effective. Introducing turbidity-inducing agents, particularly granite sand, increased Cs removal up to 37%, highlighting the role of suspended particles in enhancing coagulation. Zeolite showed improved performance due to its inherent cation exchange capacity, whereas illite and granite sand primarily contributed to turbidity formation. PB exhibited exceptional Cs removal, achieving >99.9% efficiency at 10 mg/L, with a plateau observed due to adsorption site saturation. Combining PB with coagulants did not further enhance removal but facilitated separation of PB from treated water, reducing residual contamination risks. These findings demonstrate that PB is the most promising method for Cs removal, while conventional coagulants and turbidity agents provide moderate improvements. For practical large-scale applications, future studies should focus on immobilizing PB onto stable substrates, such as filters or composite materials, to prevent leakage, simplify handling, and enable reuse. Additionally, optimization of contact time, dosage, and integration with existing treatment systems is recommended to ensure safe and effective implementation in real-world water treatment facilities.

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