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Review

Organic–Inorganic Interactions Governing Long-Term Cd Immobilization in Cement Systems Incorporating Agricultural and Metallurgical Solid Wastes: A Critical Review

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Abstract

Cadmium (Cd) is recognized as a toxic element with environmental hazards and is frequently concentrated in agricultural and metallurgical solid wastes (AgSW and MetSW). Incorporating these wastes into cement is a primary route for resource recovery. Although Cd immobilization in cement systems containing AgSW-MetSW has been examined, its mechanisms, assessment methods, and long-term stability have not been systematically synthesized from an organic–inorganic interaction perspective. This review details physicochemical characteristics and Cd occurrence forms of AgSW and MetSW, and critically evaluates Cd leaching, microstructural methods, and long-term stability. Furthermore, multidimensional immobilization pathways in AgSW-MetSW cement systems are elucidated, including physical sequestration, chemical bonding, and synergistic fixation, which exhibit significantly enhanced Cd leaching reduction in hybrid systems. The long-term stability of Cd immobilization is governed by the balance between organic matter properties and inorganic alkalinity, which determines the reaction pathways and final Cd speciation. Organic matter can immobilize Cd via complexation and pore refinement or mobilize it through soluble complexes and degradation, depending on its properties and chemical environment. Based on these findings, future priorities should include standardized leaching evaluation, in situ speciation monitoring, durability prediction under combined actions, and life-cycle cost assessment for harmless utilization of Cd.

Keywords: agricultural solid waste; metallurgical solid waste; Cd immobilization; immobilization mechanisms; organic–inorganic balance

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1. Introduction

The rapid advancement of global industrialization and agricultural modernization has been accompanied by a sharp surge in the discharge of agricultural and metallurgical solid wastes (AgSW and MetSW) [1,2]. Annual emissions reach approximately 4 billion

tonnes (dry matter) for AgSW worldwide as reported in 2023 [3], while in 2024, China's key steel enterprises alone generated 0.364 billion tonnes of MetSW comprising metallurgical slag and iron-bearing dust/sludge [4], intensifying pressure on land resources, waste management, and surrounding ecosystems. Among the contaminants associated with AgSW/MetSW streams, Cd is of particular concern because of its high toxicity, mobility, and bioavailability [5]. Once released into soil, water, or biological cycles, Cd can cause persistent damage to ecosystem functions and human health [6]. Its transfer across environmental compartments and enrichment through food chains extend the risk beyond the original disposal or storage site [7]. Given its environmental half-life of several decades, Cd pollution is cumulative, difficult to detect, and highly persistent [8]. Cd concentrations vary enormously across waste types, ranging from trace levels in agricultural residues to highly elevated levels in metallurgical dust. Incorporating Cd-bearing AgSW and MetSW into cement materials has therefore attracted attention as a route combining waste valorization with pollution control [9,10]. These wastes are integrated into concrete in diverse physical forms, assuming distinct structural functions. AgSW (e.g., Cd-enriched biomass ash) is typically utilized as a supplementary cementitious material to partially replace cement [11], whereas MetSW (e.g., slag-based materials) predominantly acts as an alkali-activated cementitious binder, which chemically and physically immobilizes heavy metals [12]. Such morphological differences and functional roles critically govern their dispersion, hydration interactions, and the ultimate Cd immobilization behavior within the cementitious matrix. Nevertheless, such utilization is environmentally defensible only when Cd is immobilized effectively and remains stable during production, service, and disposal. Reliable Cd immobilization is thus the fundamental safety prerequisite for integrating these wastes into cement recycling systems.

At present, considerable progress has been made in the immobilization of Cd in AgSW-MetSW cement systems [13–15]. While several reviews have addressed heavy-metal solidification/stabilization in cementitious and solid-waste systems [16,17], advanced Cd immobilization pathways have been explored in more recent literature. Specifically, recent review articles published in 2026 and 2027 have highlighted distinct stabilization mechanisms: the use of waste-derived geopolymers for highly efficient Cd immobilization [18], and the regulation of Cd bioavailability through coupled abiotic and microbially driven processes [19]. A systematic synthesis specifically linking the organic–inorganic interactions of AgSW and MetSW to long-term Cd immobilization, evaluation methods, and service-environment stability is still lacking. This deficiency is critical. The stability of Cd is not controlled by a single factor, but is governed jointly by waste mineralogy, cement alkalinity, hydration product evolution, and the initial chemical forms of Cd [13,17]. Meanwhile, the synergistic effects between physical sequestration and chemical bonding have not been systematically examined, nor has the long-term stability of Cd under service environments been comprehensively summarized. More importantly, laboratory observations are commonly obtained under simplified conditions that do not represent disturbances encountered during service. Carbonation, wet-dry cycles, sulfate attack, chloride ingress, and pH fluctuations can alter pore connectivity, hydration assemblages, and Cd binding states [20–23]. These changes may weaken initially effective retention pathways and create delayed release risks. A systematic synthesis for Cd immobilization is therefore needed to connect raw material characteristics, immobilization mechanisms, evaluation approaches, environmental safety, and long-term stability.

Based on this rationale, the physicochemical properties of AgSW/MetSW are systematically reviewed in this paper. Their relationships with Cd occurrence forms and environmental risks are also examined. In addition, the applicability of different methods for evaluating Cd immobilization in AgSW-MetSW cement systems, including leaching tests,

microstructural characterization, and long-term stability assessment under practical conditions, is critically compared. Particular attention is paid to three core mechanisms for Cd in AgSW-MetSW cement systems, including physical sequestration, chemical bonding, and synergistic immobilization. Physical sequestration is achieved through pore refinement, matrix densification, and spatial confinement [24]. By contrast, chemical bonding is accomplished via precipitation, surface complexation, lattice incorporation, and formation of stable Cd phases [25]. In laboratory investigations using spiked Cd, hybrid AgSW-MetSW systems have been reported to achieve a substantial reduction in Cd leaching (yielding effluent concentrations well below regulatory limits) and improved resistance to environmental disturbance.

To provide a coherent analytical framework for Cd immobilization in AgSW-MetSW cement systems, this review organizes the available evidence along a five-tier causal chain (Figure 1). On this basis, future work on AgSW-MetSW cement systems should focus on harmonized leaching evaluation, real-time speciation tracking, durability prediction under coupled loads, and life-cycle economic assessment. This review aims to deepen the scientific understanding of Cd immobilization mechanisms in AgSW-MetSW cement systems and to guide the safe, scalable, and sustainable valorization of AgSW and MetSW for ecological protection and public health.

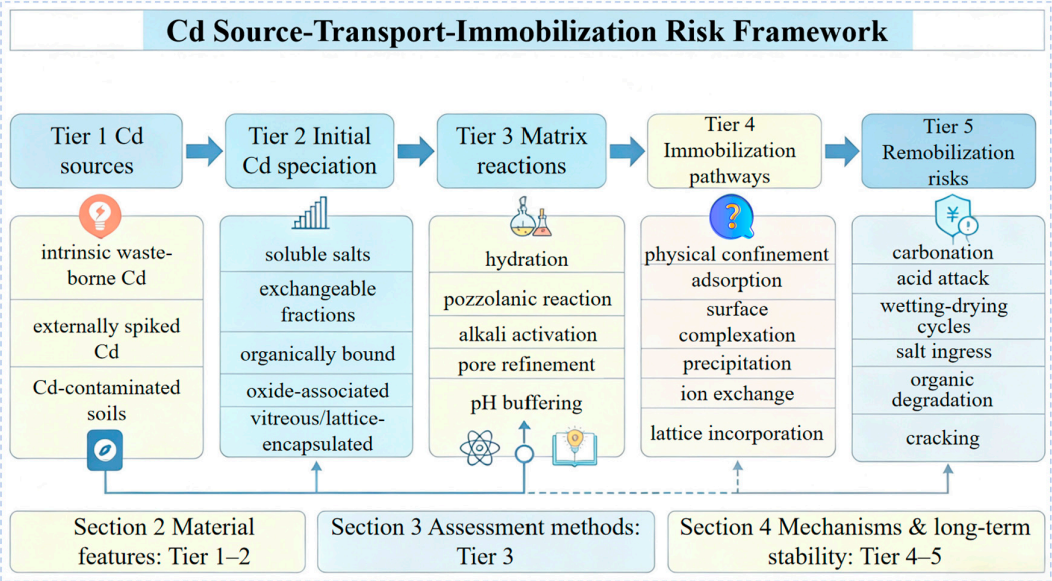


Figure 1. Five-tier causal framework for Cd immobilization in AgSW-MetSW cement systems.

The work systematically searched the Web of Science, Scopus, and Google Scholar databases for literature published between 2010 and 2026. The search strategy utilized Boolean operators to combine core terms related to (1) the target contaminant (e.g., "cadmium" OR "Cd"); (2) the treatment process (e.g., "immobilization" OR "solidification" OR "stabilization"); (3) the cementitious matrix (e.g., "cement" OR "geopolymer"); and (4) the waste type (e.g., "agricultural solid waste", "biomass ash", OR "metallurgical slag").

The screening process was conducted in two distinct steps: initially screening by titles and abstracts to exclude clearly irrelevant records, followed by a full-text review of potentially relevant articles. The inclusion criteria were (1) the article focused on Cd immobilization; (2) AgSW or MetSW were explicitly utilized as binder components or Supplementary Materials; and (3) quantitative experimental data were reported. Articles that did not report primary Cd data or lacked rigorous experimental validation were excluded. Furthermore, the methodological quality of the included publications was assessed based

on the clarity of the experimental design, completeness of reported data, and reproducibility of the results. A total of 106 articles were retained for detailed critical synthesis. The detailed bibliometric dataset extracted from the reviewed literature, including material systems, Cd sources, test methods, and quantitative results, which supports the quantitative synthesis of Cd immobilization, is provided in Supplementary Table S1.

2. Material Features of Cement Materials Incorporating AgSW and MetSW

2.1. Physicochemical Characteristics of AgSW

AgSW is defined as organic-rich agricultural residues and by-products generated in crop cultivation and livestock production, specifically including crop straws (e.g., wheat, corn, and rice straw), livestock and poultry manure, and agro-processing residues (e.g., husks, bagasse) [26,27]. Cd content and speciation differ among AgSW types (Table 1), and speciation further evolves over time. For instance, when wheat straw decomposes, Cd is initially bound and released gradually. Plant-derived Cd is mostly associated with high-molecular-weight organic matter, whereas animal-derived Cd is mainly found as inorganic Cd^{2+} and $CdOH^{+}$. Higher bioavailability is thus indicated for manure Cd.

Table 1. Cd content and speciation table of AgSW.

Category of AgSW	Cd Content (mg/kg)	Cd Speciation	References
Rice straw	-	Water-soluble Cd mainly associated with higher-molecular-weight organic matter (OM) in plant-based inputs	[28]
Wheat straw	-	Addition of wheat straw decreased exchangeable Cd and increased residual Cd during the first 5 days of decomposition; after 10 days, residual Cd decreased while exchangeable Cd increased	[29]
Peanut shell	-	As an organic amendment, peanut shell-derived biochar can reduce Cd availability	[30]
Pig manure	0.1–5.3	Water-soluble Cd primarily in inorganic aqueous forms (Cd^{2+}, $CdOH^{+}$) in animal-waste-based inputs	[28]
Poultry manure	1.3	Water-soluble Cd in animal-derived materials exists mainly as inorganic species	[28]
Cattle manures	<LOD-10	Ruminant manure has a lower proportion of OM-bound water-soluble Cd compared to non-ruminant manure	[28,31]

Note: "-" means the data are not available in the literature.

The Cd content in AgSW is significantly influenced by pollution sources and varies regionally. In unpolluted areas, Cd in straws ranges from 0.01 to 0.8 mg/kg [32], and in manure below 0.6 mg/kg [33], posing low environmental risks. Sewage-irrigated areas had serious soil Cd contamination, and 95.5% of the soil samples exceeded the screening values [34]. Cd concentrations in the livestock feed of 3.2% of swine feeds and 13.3% of beef cattle feeds exceeded the safety limit, with a maximum of 2.3 and 3.3 mg/kg, respectively [35].

Speciation of Cd determines the environmental risk of Cd. According to the procedures for sequential extraction [36,37], Cd in AgSW can be divided into five fractions: exchangeable, carbonate-bound, Fe-Mn oxide-bound, organic-bound, and residual [38].

Among them, the organic-bound fraction is generally a relatively large proportion because straw-derived organic matter has strong chelating affinity for Cd [39]. The residual fraction is sequestered in the crystal lattice and has very low mobility [37]. The exchangeable and carbonate-bound fractions are generally more easily affected by environmental changes, such as acid conditions or anaerobic environments [36,38].

Although organic matter in AgSW provides functional groups for Cd complexation and pore modification, it is not always advantageous. Low-molecular-weight dissolved organic matter can form soluble Cd complexes to increase the mobility of Cd under alkaline conditions [39]. Carbohydrates and organic acids may inhibit cement hydration by adsorbing cement particles and chelating Ca^{2+}, thus delaying setting and impairing matrix densification [40]. Long-term stability is further challenged by the biodegradable nature of AgSW organics. Microbial decomposition releases CO_2, lowers pH, and opens up new pores, all of which can remobilize Cd that was previously immobilized [41]. Livestock manure also contains Cl^- and NH_4^+ that compete for binding sites and increase the solubility of Cd by complexation [8,42]. The two paths suggest that organic matter is not a general immobilizer of Cd; its net contribution varies depending on factors such as molecular size, degradability and the local alkalinity environment [39]. Moreover, industrial utilization of AgSW usually requires thermal pretreatment (combustion or pyrolysis), which drastically alters Cd speciation. For instance, Cd in straw ash is highly water-leachable (as soluble $CdCl_2$), whereas wood ash binds Cd more stably as oxides or $CdSiO_3$ [43]; meanwhile, elevated ashing temperatures (600–1000 °C) promote Cd volatilization and reduce residual Cd in the ash [44]. Consequently, ash-derived Cd is not necessarily less labile than raw biomass Cd, and its immobilization in cement systems ultimately depends on ash mineralogy, thermal history, and matrix alkalinity.

2.2. Physicochemical Characteristics of MetSW

MetSW is the solid waste generated in metal smelting. It mainly refers to slag from ferrous and non-ferrous metal production [45]. Steel slag, copper slag, lead-zinc slag, red mud, and nickel slag are examples [46]. Cd is also a heavy metal that occurs in these sources [47]. These slags are noted for their high inorganic mineral content and variable particle sizes. They are irregularly shaped particles or lumps with different colors due to different core components. The density of these slags ranges from 3.6 to 3.9 kg/m^3, depending on mineral composition [48,49]. Particle sizes of these materials vary widely. Coarse particles are as large as tens of centimeters, and fine particles are on the order of micrometers [49].

Table 2 summarizes the Cd contents from 2.19 to 14,000 mg/kg and the speciation in MetSW and related contaminated media. In primary MetSW, Cd in copper dust exists as soluble salts with high mobility, whereas in slag it is fixed in stable glassy phases. By contrast, smelting-affected environmental media such as waste-deposit soils, coke grains, and alloy fragments exhibit high Cd contents up to thousands of mg/kg and specific speciation including solid solutions or fragments. These media are closely linked to smelting-related environmental risks. Therefore, the total Cd concentration alone is a poor predictor of risk, as the hazard is ultimately governed by its speciation.

Table 2. Cd content and speciation in MetSW and related contaminated media.

Category	Cd Content (mg/kg)	Cd Speciation	References
MetSW			
Copper smelting ESP converter dust	5400	Cd may exist in the amorphous phase as soluble salts	[50]
Slag	14–14,000	Cd hosted in slag glass and vitreous phases	[51]
Cd-contaminated Environmental Media			
Pb-Zn metallurgical waste deposit soil	2.19–49.84	BCR sequential extraction: Cd in mobile fractions 34.7%	[52]
Coke grains	Up to 2700	Cd exhibits metal enrichment zones at grain edges	[53]
Multimetallic oxides (Sn, Fe, Pb, Zn)	Up to 5700	Cd is present in multimetallic oxide phases	[53]
Multimetallic alloys	Up to 7700	Cd occurring in alloy fragments with dimensions of a few microns	[53]

3. Research Methods for Evaluating Cd Immobilization in Cement Materials Incorporating AgSW and MetSW

Before detailing specific leaching and characterization methods, it is essential to distinguish three fundamentally different Cd immobilization systems. In the first, Cd is intrinsically hosted in AgSW/MetSW (e.g., smelting dust, slag, or contaminated straw), where its initial speciation, host-phase stability under alkali activation, and interference by coexisting ions (Zn, Pb, Cl^-, SO_4^{2-}) regulate the release [41,54]. In the second, soluble Cd salts are added to cement pastes and immobilized by binders, but the labile nature of these salts may lead to an overestimation of immobilization efficiency [55]. In the third, Cd is spiked into the soil and treated with the same binders, but organic matter and clay in the soil may have interfered with Cd retention, thus complicating the attribution to binder effects alone [56]. To enable rigorous cross-literature comparison, the evidence is divided into three categories: Type I—intrinsic Cd-bearing waste; Type II—Cd-spiked cement systems; and Type III—Cd-spiked soil immobilized by waste-derived binders.

3.1. TCLP Leaching Test for Cd Release and Immobilization Efficiency

The Toxicity Characteristic Leaching Procedure (TCLP) is conducted in an acetic acid buffer (pH 4.93 ± 0.05) to simulate acidic landfill leachate. Samples are mixed at a 1:20 solid-to-liquid ratio and shaken at 25 ± 2 °C for 18 ± 2 h. TCLP is widely used in regulatory applications, but it is limited to only an acetic acid buffer, and alkaline cement matrices or neutral groundwater are not included. Another shortcoming is that the short-term batch test cannot account for long-term aging, microbial activity, and organic matter degradation [57]. Therefore, TCLP results alone are not considered sufficient, and complementary protocols are recommended to cover a broader range of environmental conditions. To address these limitations, TCLP has been combined with sequential extraction data in several reviewed publications. The combined method connects the large-scale leaching behavior to microscale changes in speciation and thus provides mechanistic insight into the reduction in Cd leaching.

For example, the concentration and percentage leaching of Cd from sludge containing MetSW under anoxic and oxic conditions during the TCLP leaching test (Type I) have been documented [58]. The leaching of Cd is found to be lower under oxic conditions, which is attributed to immobilization by iron oxide precipitates formed during the test. In Type II cement systems, Cd-bearing powders were immobilized by binders in cement pastes. Leaching tests with TCLP under simulated silo conditions showed Cd to be the least leachable metal, predominantly retained within the matrix [59]. In addition, Figure 2 [60] presents a soil remediation system, where the observed decline in TCLP-extractable Cd (from ~0.33 to ~0.20 mg/kg) primarily reflects physical entrapment and surface complexation within the soil matrix, rather than cement hydration effects. This case provides a relevant analogue for Type III systems and, more importantly, highlights a methodological limitation: TCLP alone cannot fully distinguish between cementitious binding and non-cement physical sequestration, reinforcing the need for complementary microstructural characterization.

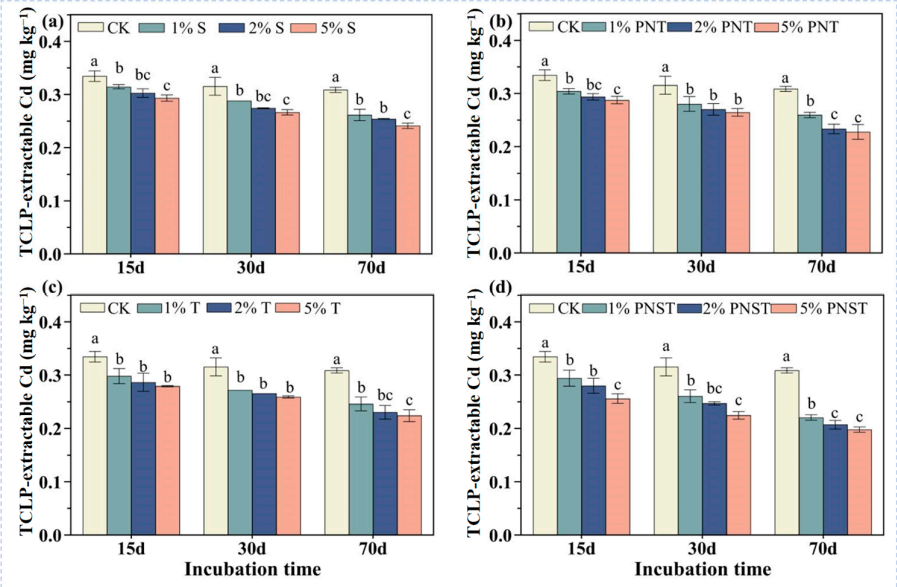


Figure 2. Soil TCLP-extractable Cd concentration in a Cd-contaminated soil system treated with (**a**) nanosilica, (**b**) PVA/SA/N-TM, (**c**) tourmaline, and (**d**) PVA/SA/N-silica@TM during the incubation. Different letters (a, b, c, bc) above the error bars indicate significant differences among treatments at $p < 0.05$; treatments sharing a common letter (e.g., bc and c) are not significantly different, whereas those with no shared letter (e.g., a and b) are significantly different [60]. (Reproduced with permission from Xu et al., Environmental Technology & Innovation; published by Elsevier, 2025).

The TCLP test has been established as a key method for evaluating Cd immobilization by simulating acidic landfill environments. However, a serious deficiency is the lack of standardized testing protocols for AgSW-MetSW cement systems: variations in sample preparation, solid-to-liquid ratio, and leachant composition complicate direct comparisons among different works. Future standardization will focus on system-specific protocols for more rigorous assessments.

3.2. Microscopic Analysis for Cd Distribution and Matrix Densification

Microstructure analysis reveals the spatial distribution of Cd and the densification evolution of the cement matrix by characterizing micro-morphology, phase composition, and elemental associations within the system [21,25,61].

For instance, a specific SEM-EDS paper [62] showed that Cd accounted for 0.6% of the normalized elemental mass in the newly formed aggregates, and elemental mapping further confirmed the spatial co-localization of Cd with Ca, Fe, and O. This co-localization pattern is regarded as evidence of Cd coprecipitation with Ca-Fe-bearing phases. Similarly, XRD patterns reported in the literature [63] showed discrete crystalline phases, including C-S-H, N-A-S-H, $C_3Al_2Si_2O_8$, and $Cd(OH)_2$, with the latter directly indicating that Cd had precipitated as a separate hydroxide phase. In addition, the broad humps centered at Cd-2 and Cd-4 positions suggest Cd substitution into the lattice of C-A-S-H, reflecting structural incorporation. Complementing these XRD observations, reported FTIR spectra [63] also provided direct vibrational information about the local coordination environment of Cd in the reaction products. Perturbation of the Si-O and Al-O stretching modes showed that Si-O-Cd and Al-O-Cd bonds had formed, and thus the chemical environment around Cd sites was altered. Based on these microstructural observations, the evidence reviewed points to three main mechanisms that together control Cd retention: surface complexation on C-A-S-H surfaces, precipitation as Cd-hydroxide/carbonate, and structural incorporation into silicate hydrate frameworks [16].

Furthermore, TG-DTG thermograms from a previous work [64] showed two distinct endothermic peaks at 701.69 °C (T3) and 772.79 °C (T4). These high-temperature events primarily correspond to the decomposition of the protective carbon layer and the structural breakdown of the aluminosilicate matrix. Nevertheless, Cd is effectively immobilized in the matrix. Overall, different papers report varying FTIR band intensities and TG endothermic peaks, indicating that the relative contributions of these pathways depend on waste composition, curing age, and system alkalinity [65]. This mechanistic multiplicity goes beyond the previous assumption of precipitation-dominated retention and implies that formulation optimization requires a tailored approach accounting for the specific organic–inorganic balance.

Microscopic analysis of the AgSW-MetSW cement system shows that Cd immobilization is due to both chemical bonding (precipitation, co-crystallization, and complexation) and physical sequestration through matrix densification. Pore refinement and phase assembly densify the matrix, which restricts ion diffusion, lowers permeability, and thereby curtails Cd mobility [66]. However, conventional ex situ characterization cannot show the dynamic interfacial reaction, and spatial heterogeneity of Cd poses a sampling problem [67]. In the reviewed literature, ex situ analysis has been exclusively applied to aged samples, capturing only a single fixed endpoint. Future developments will need in situ or operando characterization to monitor changes in Cd speciation and microstructure in real time.

3.3. Long-Term Performance Assessment of Cd Immobilization Stability

A stability verification system for long-term Cd immobilization performance is established based on kinetic fitting and accelerated leaching tests. To investigate the immobilization kinetics, the short-term data [68] are fitted to a pseudo-second-order kinetic model [Equation (1)]

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e}, \quad (1)$$

where q_t and q_e are the amounts of Cd immobilized at time t and at equilibrium (mg/kg), respectively, and k_2 is the rate constant of the pseudo-second-order model. Concurrently, the effective diffusion coefficient (De) is calculated according to the ANS 16.1 formulation [Equation (2)]

$$D_e = \pi \left[\frac{a_n/A_0}{(\Delta t)_n}\right]^2 \left[\frac{V}{S}\right]^2 T, \quad (2)$$

where a_n is the amount of Cd leached during interval n, A_0 is the total amount of Cd in the specimen, V is the volume of the specimen, S is the geometric surface area, $(\Delta t)_n$ is the duration of interval n, and T is the mean leaching time. A lower De value indicates stronger resistance to Cd migration and thus superior long-term stability.

Beyond kinetic modeling, empirical insights into long-term stability are provided by sequential extraction data from field-aged soils. As depicted in Figure 3a [69], the residual Cd fraction exhibited a substantial increase across all treatments from the short-term (6 months) to the long-term (12 years), with the ATP treatment yielding the highest residual fraction and the lowest exchangeable fraction. Corroborating this trend, Figure 3b [69] reveals the absolute concentrations of Cd fractions in soil colloids, where the ATP treatment sustained the highest levels of residual and crystalline iron oxide-bound Cd after 12 years, underscoring its superior long-term immobilization efficacy. Collectively, these long-term field observations demonstrate the progressive transformation of Cd into highly stable fractions, thereby corroborating the sustained stability of the immobilization performance.

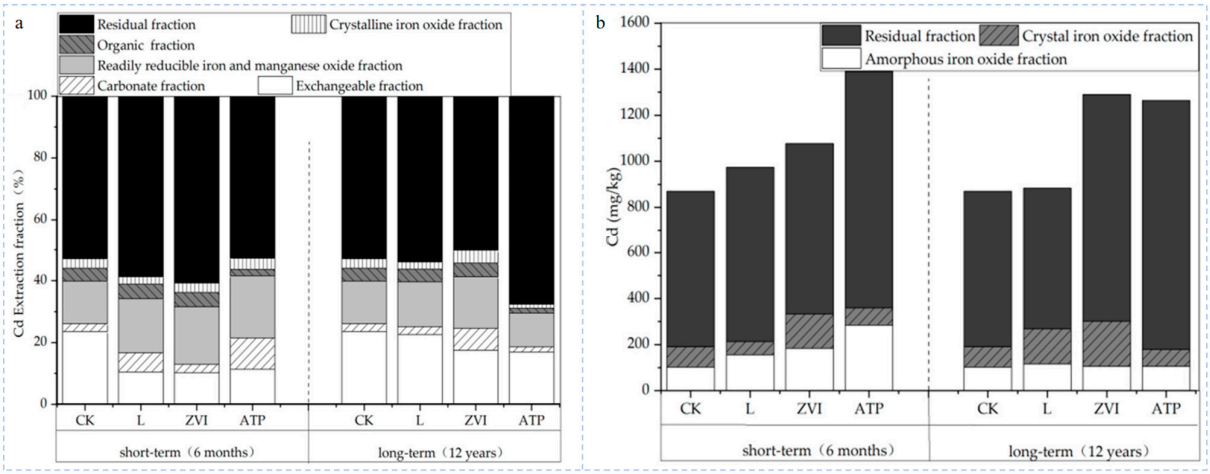


Figure 3. The long-term immobilization performance and stability of Cd in soil [69]: (**a**) The fraction distribution of soil Cd in short/long term. (**b**) The fraction concentration of soil colloid Cd in short/long term. CK: control; L: lime; ZVI: zero-valent iron; ATP: attapulgite. (Reproduced with permission from Liu et al., Toxics; published by MDPI, 2024).

Limited long-term insight is provided by these empirical fittings, because long-term Cd stability is governed by coupled environmental actions by which phase composition, pore structure, and chemical microenvironment are progressively altered [70,71]. Carbonation is initiated by CO_2 ingress, by which $Ca(OH)_2$ is neutralized and C-S-H is decalcified. The consequent pH drop may cause $Cd(OH)_2$ precipitates to be dissolved, while Cd is reprecipitated as $CdCO_3$ by simultaneously generated carbonate ions, and the net effect is rendered condition-dependent [72]. Acid attack is driven by external H^+; hydration products are dissolved and porosity is increased, whereby binding sites are destroyed and Cd diffusion is facilitated [73]. Repeated shrinkage and expansion are induced by wet-dry cycles, and microcracks are generated, by which transport pathways are shortened and physically sequestered Cd is released [74]. Sulfate and chloride ingress in the AgSW-MetSW cement matrix will reduce Cd stability by forming expansive ettringite (causing cracks that weaken physical entrapment) and competing for sorption sites [75]. These paths make the short-term leaching insufficient for field predictions; therefore, accelerated ageing coupled with mechanistic modelling is required to assess delayed Cd remobilization.

In the AgSW-MetSW cement system, accelerated tests are used to simulate extreme environmental conditions and assess the long-term performance of Cd immobilization. Dry-wet cycles are used to simulate seasonal wet and dry alternation, and changes in Cd leaching and crack formation are monitored to evaluate the pore-filling effect [76]. Given the inherent heterogeneity of AgSW and MetSW, crack growth and pore connections are affected by cyclic wet-dry cycles, which in turn weakens Cd physical confinement [77]. Cold-region temperature swings are reproduced by freeze–thaw cycling, and the resulting freeze resistance is evaluated through compressive strength tests and XRD, because the microcracks that form can undermine the physical entrapment of Cd. The complicated phase assemblage of this matrix may have differential freeze–thaw responses and thus create preferential pathways for Cd release [78]. By simulating thermal exposure, high-temperature aging can induce bond breakage that, as tracked by FTIR, weakens Cd complexation. Organic matter in AgSW and metastable phases of MetSW may decompose or change under high-temperature treatment to release immobilized Cd [79]. When these stresses act together, they can drive both physical and chemical remobilization of Cd. Reliably evaluating long-term retention stability therefore hinges on combining pore structure, elemental distribution, and speciation data. The extended Cd-immobilization capacity is confirmed when the difference between the model prediction and the accelerated test extrapolation falls within the acceptable range. These findings confirm that the AgSW-MetSW cement matrix can maintain structural integrity and chemical fixation under engineering conditions.

4. Cd Immobilization Mechanisms in Cement Materials Incorporating AgSW and MetSW

4.1. Physical Sequestration of Cd Through Pore Refinement and Matrix Sequestration

Physical sequestration constitutes a primary barrier governing Cd mobility by restricting diffusion and access to reactive sites [24]. In the AgSW-MetSW cement system, this mechanism is realized through pore refinement and matrix densification [80], yet its manifestation varies across different material systems. As shown in a representative metal-oxide system (Figure 4 [81]), Cd can be physically confined within the structural interstices of layered materials. A comparable spatial confinement pathway is expected to occur at the interfaces of cement hydration products, where Cd is physically trapped within the dense matrix. In addition, in aluminosilicate-based geopolymers, Cd is physically sequestered within the three-dimensional network [63].

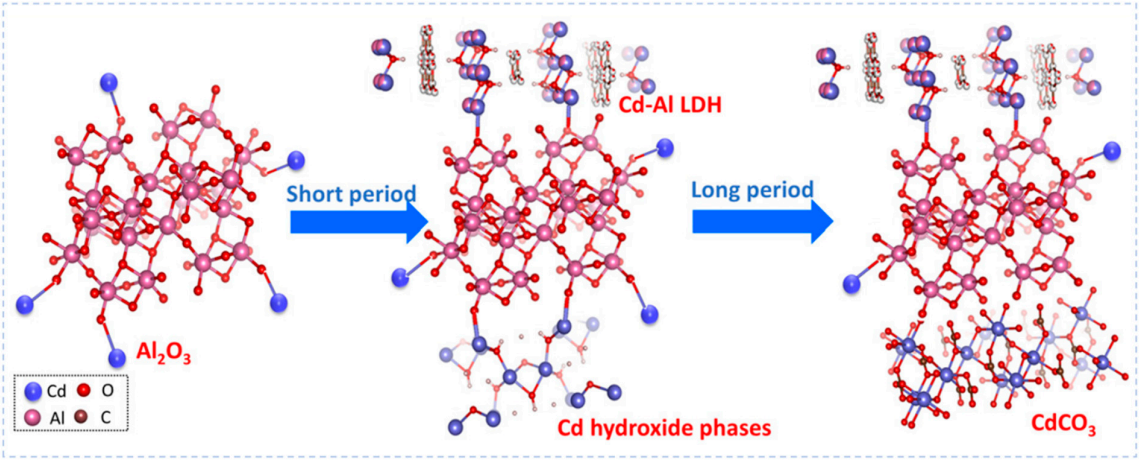


Figure 4. Molecular mechanism of Cd adsorption and immobilization on metal oxide surfaces [81], illustrating the general surface complexation pathway that is also relevant to Cd retention on cement

hydration products. (Reproduced with permission from Sun et al., Environment International; published by Elsevier, 2019).

The same core principle in AgSW-MetSW cement focuses on the dense microstructure formed by hydration [82,83]. It is realized through a multi-level synergy that integrates three complementary aspects, avoiding reliance on any one isolated pathway. A three-dimensional network is generated by hydration as the first level [84]. Cd and particles are mechanically sequestered within it. Short-term physical entrapment is provided by the layered interlayer spaces [67]. Long-term fixation is achieved through carbonate lattice incorporation. Network densification is promoted by active components from AgSW and MetSW [85,86]. Secondly, pore structure is optimized by micron-scale fibers from AgSW. Large pores are blocked and pore sizes are refined. Diffusion paths are extended and permeability is reduced [87]. The matrix is also toughened, inhibiting microcracks induced by wet-dry cycles. The third level is that hydration pores are filled by MetSW particles. Fine powders and C-S-H gel jointly densify the interfacial transition zone, creating a particle-gel compaction effect [88], lowering the connectivity of pores and reducing convective paths. Thus, the migration of Cd is effectively suppressed. Acting together, these three mechanisms generate a dense physical barrier that increases matrix compactness and substantially curtails Cd leaching.

4.2. Chemical Bonding of Cd Through Precipitation, Complexation, and Ion Exchange

In AgSW-MetSW cement systems, chemical bonding governs the speciation and long-term stability of immobilized Cd through precipitation, complexation, and ion exchange [89,90]. The above reactions produce stable Cd-bearing phases that can be used for long-term immobilization. Integrating the underlying physicochemical principles reveals the essential functional processes.

For the system containing red mud, blast furnace slag, calcium carbide residue, and phosphogypsum (Type III) [65], hydration and pozzolanic reactions produce C-A-S-H gel and AFt. Soil pores are filled by these products. Cd is immobilized stably through surface complexation, ion exchange, and co-precipitation. Surface complexation is accomplished by Cd bonding with hydroxyl groups. Ion exchange is realized when Ca^{2+} is replaced by Cd. Cd-bearing phases are formed through co-precipitation. For the RAC geopolymer system (Type III) [62], Si-O and Al-O bonds are broken and reorganized. N-A-S-H and C-A-S-H gels are formed. Compactness is enhanced by these gels through pore filling. Carriers are provided for Cd co-precipitation. Long-term performance of $Cd(OH)_2$ precipitates is maintained by the highly alkaline environment. At the lattice scale (Figure 5 [91]), Cd^{2+} can directly substitute the Ca^{2+} in the octahedral layers of CaFe layered double hydroxide (CaFe-LDH). This isomorphic replacement mechanism is analogous to the substitution of Ca^{2+} by Cd^{2+} within the C-S-H or AFt structures in cement matrices. Concurrently, surface complexation occurs as Cd^{2+} bonds with hydroxyl groups at the edges of the layers, synergistically contributing to the long-term immobilization and enhanced stability of Cd.

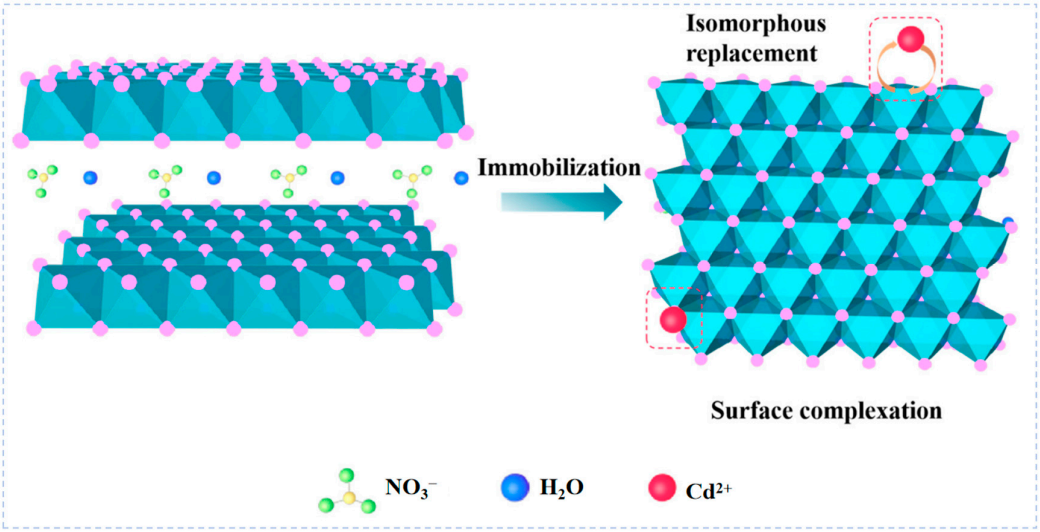


Figure 5. Schematic illustration of Cd immobilization on CaFe-LDH [91], showing isomorphic substitution and surface complexation pathways analogous to those occurring in Cd-bearing cement hydration products. (Reproduced with permission from Jing et al., *Minerals*; published by MDPI, 2024).

Overall, co-precipitation, complexation, and ion exchange are used to convert Cd into insoluble crystals or stable organometallic complexes [92]. A synergistic effect is exerted by this chemical barrier in the alkaline hydration environment. Extremely low solubility and strong binding capacity are possessed by it. Long-term Cd immobilization is achieved.

4.3. Synergistic Immobilization of Cd Through Coupled Physical Sequestration and Chemical Bonding

In cement systems prepared from AgSW-MetSW, Cd immobilization is not achieved by physical sequestration or chemical bonding alone. Instead, a synergistic cycle driven by the coupling of structural entrapment and chemical reactions promotes the conversion of Cd from labile to stable forms [93]. This synergistic effect is visually depicted in Figure 6a [93], where the physical sequestration of hydrated Cd ions is coupled with chemical bonding mechanisms, including Cd-matrix association with Si-O and Al-O environments, cation exchange, and precipitation within the silicate/aluminate framework. Chemical bonding and physical sequestration are mutually dependent in this cycle to enhance Cd immobilization. This synergistic immobilization mechanism is also observed in non-cement materials. Similar synergistic pathways are illustrated in Figure 6b [94], using a gel-bead soil remediation system as a macroscopic analogue for the coupling of physical retention and chemical binding. In this system, the material achieves efficient physical sequestration through water retention and pore filling. At the chemical level, surface adsorption and isomorphic substitution effectively bind Cd, while the sustained release of urea provides nutrients (C, N) that facilitate vegetation growth, indirectly enhancing the rhizosphere stabilization of Cd. Furthermore, the magnetic properties of the material allow for magnetic recovery, ensuring its recyclability in practical applications.

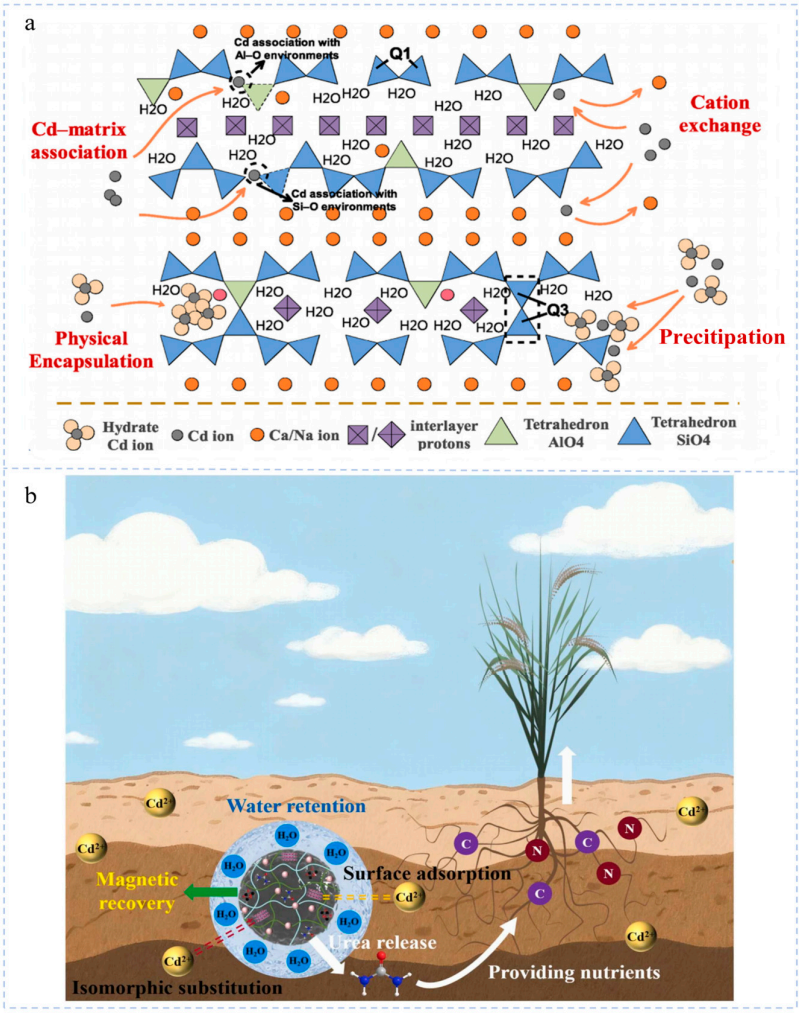


Figure 6. Synergistic physico-chemical immobilization mechanisms of Cd: (**a**) Micro-scale mechanisms in the cementitious matrix: physical sequestration coupled with chemical bonding (Cd-matrix association, ion exchange, and precipitation) [93]. (Reproduced with permission from Wang, Journal of Hazardous Materials Advances; published by Elsevier, 2026.) (**b**) Macro-scale mechanisms in soil remediation: physical sequestration coupled with chemical complexation (surface adsorption, isomorphic substitution) [94], illustrating the general coupling of physical and chemical retention pathways. (Reproduced with permission from Zhao et al., Green and Smart Mining Engineering; published by KeAi, 2026).

The way bridging complexes and hydration products work together highlights how closely physical sequestration and chemical bonding are linked in Cd immobilization. Specifically, humic substances form bridging complexes via Ca^{2+}/Fe^{3+}, and N-ligands establish stable chemical bonds with Cd, thereby tightly binding Cd to hydration products and further sequestering it in C-S-H gel to inhibit its migration [55,61]. Fibrous organic matter refines the pores, an effect that not only tightens physical confinement but also supplies rich adsorption interfaces. At the same time, the organic matter adsorbed on C-S-H raises the density of active sites, which further increases the sorption capacity for Cd [95].

At the same time, inorganic hydration phases ($Ca(OH)_2$ and AFt) immobilize Cd by ion exchange and precipitation reactions, and these phases are encapsulated within the C-S-H network; thus, they are protected from external attack and the overall matrix stability is maintained through bonding products [21]. However, it should be pointed out that the

durability of this protective layer may also be affected by environmental factors such as carbonation and sulfate attack, and its long-term stability has not yet been verified through organized environmental ageing experiments [20,96].

The above mechanisms have been verified by experiments in many cement systems. As shown in Table 3, the five binder systems based on AgSW and MetSW have moved from a single-mechanism model to a synergistic physical-chemical coupling system. Straw biochar cement (Type I) reduces the intrinsic Cd leaching by 30.1%–49.6% through pore refinement and organic complexation [97], but its performance varies according to the feedstock and has only been verified up to 28 days. Alkali-activated red mud slag (Type I/II) and geopolymers (Type II) mainly achieve immobilization through the incorporation of N-A-S-H gel or charge compensation, with immobilization rates of 97% [78] and 99% [84], respectively. However, the spiked $Cd(NO_3)_2$ in the geopolymers is significantly more mobile than the intrinsic Cd, and pH-dependent leaching of red mud shows instability under varying acidity. By contrast, hybrid AgSW-MetSW systems (Type II/III) intentionally couple physical pore filling with chemical bonding. The combined mechanism shows a good reduction of 70%–90% in Cd leaching over 7–56 days of curing and keeps the effluent Cd below 1.0 mg/L. A similar synergy occurs in OPC-blended (Type III) systems, where C-S-H binding and coprecipitation achieve a 72.3% [98] reduction in Cd in real contaminated soil with TCLP leachate below 1.0 mg/L. That review summarized the key evidence and limitations of each system, revealing that Cd leaching, long-term stability, and microstructure are linked through the synergy of physical sequestration and chemical bonding. Available evidence indicates that properly designed hybrid systems can offer complementary advantages, but the effectiveness of their synergy is strictly governed by the proportional balance between organic and inorganic components [55]. Leaching reduction quantifies the decrease in Cd concentration in the leachate relative to the untreated material, whereas immobilization rate reflects the proportion of Cd retained in the solid matrix; because these two metrics rest on different evaluation bases, direct cross-comparisons among these systems remain relatively limited [99], and the general rules for optimal ratios and material types still require more systematic investigation.

Table 3. Comparative summary of Cd immobilization research in AgSW-MetSW cement systems.

Attribute	AgSW (Straw Biochar)	MetSW (Red Mud)	Geopolymer	Hybrid AgSW-MetSW	OPC
Type	I	I/II	II	II/III	III
Initial Cd loading	Intrinsic: straw-borne, 0.481–3.520 mg/kg	Intrinsic (oxide) + spiked in some cases	Spiked $Cd(NO_3)_2$	Intrinsic + spiked $Cd(NO_3)_2$ or soil	Cd-contaminated soil
Binder composition	Cement + biochar (5 wt%)	Alkali-activated red mud + slag	Fly ash/slag-based geopolymer	AgSW + MetSW + cement (variable)	OPC-based binder
Curing age	7–28 d	7–28 d	7–28 d	7–56 d	-
Leaching method	TCLP	TCLP/pH-dependent	TCLP/semi-dynamic	TCLP/sequential extraction	TCLP
Leachate pH	4.93	4.93/2–12	4.93/variable	4.93	4.93
Cd concentration	-	≤0.21 mg/L	-	<1.0 mg/L	<1.0 mg/L

Leaching reduction	30.1–49.6%	-	-	70–90% (estimated)	72.3% (with 5 wt% OPC)
Immobilization rate	-	>97%	>99%	-	-
Main evidence	Pore refinement; complexation with organic functional groups	N-A-S-H gel incorporation; pH buffering	N-A-S-H charge compensation; physical sequestration	Complementary: pore refinement + chemical bonding	C-S-H binding; co-precipitation
Limitations	Biochar variability; short-term only; no field validation	High alkalinity; coexisting metals interfere	Spiked Cd far more mobile than real waste; lab-scale only	Condition-dependent synergy; spiked Cd is more labile; long-term evidence scarce	Soil matrix complexity; competing ions (Cu^{2+}, Pb^{2+})
References	[97,100]	[62,78,101]	[63,84,102]	[25,103–105]	[13,98]

Note: "-" means the data are not available in the literature.

To move beyond a descriptive summary, the data in Table 3 were cross-examined to construct an original quantitative synthesis. A distinct performance hierarchy emerges: single AgSW systems exhibit the lowest leaching reduction, whereas hybrid AgSW-MetSW cement systems show a marked non-linear enhancement. This transition indicates that the synergy between organic and inorganic components is complementary rather than merely additive. However, a critical divergence arises between the high immobilization rates reported for alkali-activated cement systems and their actual environmental stability. Because these rates are predominantly derived from spiked $Cd(NO_3)_2$ systems, in which soluble Cd is highly labile, they may overestimate long-term retention relative to the intrinsic Cd in hybrid cement systems. This cross-comparison suggests that the apparent superiority of alkali-activated cement systems may reflect differences in test conditions rather than intrinsic matrix stability. Accordingly, the leaching reduction observed in hybrid cement systems provides a more reliable and realistic benchmark for the valorization of intrinsic Cd-bearing wastes.

In short, the distribution of Cd is limited by both chemical bonding and physical sequestration; the former restricts the speciation of Cd, and the latter reduces the space available for migration [106]. The synergy of these two mechanisms stably retains the vast majority of residual Cd, thus achieving long-term immobilization of Cd [13]. However, the present conclusions are mainly based on laboratory-scale and short-term leaching experiments. Whether they can be applied in complex hydrogeological conditions at the field site and whether they are suitable for various pollution loads still need to be verified by in situ monitoring and long-term observation.

5. Conclusions and Prospects

5.1. Conclusions

This review systematically examined the physicochemical features, evaluation methods, and immobilization mechanisms of Cd in AgSW-MetSW cement systems. Building on the evidence synthesized above, the key conclusions are drawn as follows:

(1) In AgSW-MetSW cement systems, three synergistic mechanisms for Cd immobilization are identified: physical sequestration by hydration product networks, chemical

bonding via surface complexation, ion exchange, and coprecipitation, and their coupled interplay. Based on these coupled pathways, hybrid formulations have been reported to achieve a substantial reduction in Cd leaching (yielding effluent concentrations well below regulatory limits) in spiked-Cd laboratory experiments. The net effect of these pathways is dependent on conditions, governed by organic composition, alkalinity, and hydration progress.

(2) The evaluation of Cd immobilization efficacy in AgSW-MetSW cement systems using multiple approaches is summarized: TCLP leaching tests for environmental safety, microstructure, phase composition, chemical bonding, and thermal stability phase composition for microstructural characterization, and kinetic modeling combined with accelerated environmental simulations for long-term performance projection.

(3) The control of long-term leachability reduction in Cd by hydration products in AgSW-MetSW-based systems is outlined: C-S-H gel provides high-capacity sorption and structural incorporation; AFt phase enables lattice substitution and pore refinement; geopolymer networks offer charge neutralization and acid-resistant sequestration. Microscopic elemental analyses reported in the reviewed literature indicate that Cd accounts for a minor fraction (typically below ~1%) of the elemental mass, confirming its stable retention within the hydration products.

(4) The long-term stability of Cd immobilization is governed primarily by the balance between organic matter properties and inorganic phase alkalinity, which dictates reaction pathways and phase evolution. Organic matter acts as a double-edged sword, enhancing Cd retention via complexation and pore refinement, but promoting mobilization when labile low-molecular-weight organics dominate. This balance ultimately determines the final Cd speciation and its environmental release risk.

5.2. Prospects

The large-scale application of Cd immobilization technologies remains hindered by insufficient validation of durability under carbonation, acid attack, and wet-dry cycles, absence of regulatory frameworks, high costs from binder consumption and logistics, and feedstock heterogeneity. Future research should address four interconnected priorities. A standardized leaching evaluation framework specifically for AgSW-MetSW cement systems is urgently needed, as current protocol variations impede direct comparability and harmonized procedures are essential for regulatory acceptance. Advanced in situ or operando spectroscopic characterization should be employed to obtain direct evidence of Cd binding states, tracking its dynamic speciation evolution during hydration and environmental exposure, since ex situ endpoint analyses cannot capture transient interfacial reactions governing long-term retention. Durability experiments under coupled actions including carbonation, sulfate attack, chloride ingress, and wet-dry cycles are required to quantify Cd remobilization under realistic service conditions and to develop predictive models from multi-factor data. Life cycle assessment and cost analysis must be integrated to evaluate genuine environmental benefits and elucidate trade-offs among binder consumption, immobilization efficiency, and durability.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/min16111152/s1. Table S1. Bibliometric dataset of Cd immobilization performance extracted from the reviewed literature. The quantitative basis for the statements in the main text is provided by this dataset, in which material systems, Cd sources, test methods, results, and corresponding mechanisms are detailed.

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draft, visualization. M.C.: resources, supervision. X.L. (Xiaoming Liu): writing—original draft, writing—review and editing. Y.L.: supervision, writing—original draft. Y.W.: writing—review and editing, supervision. All authors have read and agreed to the published version of the manuscript.

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