



## The fate of heavy metals in recycled concrete paste upon enforced carbonation: A review

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### ABSTRACT

Enforced carbonation of recycled concrete paste (RCP) presents a promising solution for carbon capture and utilization (CCU) in the cement and construction industries. By transforming concrete waste into a reactive pozzolan while sequestering CO<sub>2</sub>, enforced carbonation addresses major environmental challenges and aligns with the circular economy concept in the cement and concrete sector. This review contextualizes the impact of enforced carbonation on heavy metals in RCP to ensure sustainable and ecological compatibility of both the process and the final product, carbonated RCP (cRCP). It summarizes current knowledge on heavy metals in cement and proposes hypotheses on their plausible immobilization mechanisms in enforced-carbonated materials. The review highlights gaps in the literature concerning the impact of carbonation on heavy metals in cement, underscoring the necessity of exploring novel approaches for assessing the heavy metal behaviors from RCP to cRCP.

### 1. Introduction and context

As the importance of sustainability is heavily promoted in the construction industry, recycling and valorization of construction and demolition waste (CDW) is increasingly vital (Oh et al., 2021; Schneider, 2019). Demolished concrete from the End-of-Life (EoL) constructions, comprising up to 75 % of demolition waste (Coelho and De Brito, 2011), accounts for a significant portion of the estimated 350 million tons of CDW generated annually in Europe (Gebremariam et al., 2020).

Meanwhile, the reduction of CO<sub>2</sub> emissions remains the most pressing global challenge within construction sector. In the cement industry, clinker production emission alone contributes approximately 7 % of global anthropogenic CO<sub>2</sub> emissions (Schneider, 2019). In this context, recycled concrete paste (RCP), derived from the fine fraction of demolished concrete via smart separation technology, has emerged as a promising CDW source material for CO<sub>2</sub> sequestration and circular applications (Driver et al., 2024; Fang et al., 2021; Katsuyama et al., 2005; Zhao et al., 2024). RCP can be treated with enforced carbonation, an engineered form of accelerated mineral carbonation techniques, which transforms cementitious materials into stable calcium carbonate (CaCO<sub>3</sub>) and reactive pozzolan (Snellings and Matschei, 2025; Zajac et al., 2023b; Zhang et al., 2019). Enforced carbonation not only enables

permanent CO<sub>2</sub> storage, but also effectively valorizes RCP into reactive pozzolan, which has the potential to replace the clinker content in cement (Fig. 1) (Li et al., 2022; Mao et al., 2023; Poon et al., 2023; Zajac et al., 2021b; Zhang et al., 2019). The incorporation of cRCP as an SCM allows partial replacement of clinker content in cement, further reducing the CO<sub>2</sub> footprint associated with cement production (Liu et al., 2025; Teune et al., 2023). This circular strategy offers a dual environmental benefit, addressing both CDW valorization and carbon reduction within the cement and concrete value chain.

While fully carbonated RCP has demonstrated excellent pozzolanic properties and promising prospect as an SCM, the environmental compatibility of the process and materials depends on managing the behavior of potentially hazardous heavy metals present in RCP. The aqueous environment during enforced carbonation accelerates mineralization but also influences the mobility and fate of heavy metals. Therefore, assessing their speciation, leachability, and immobilization during enforced carbonation is critical to ensuring the sustainability and safety of recycled concrete materials. Yet, to date, little is known about how enforced carbonation affects the heavy metals in cRCP. This review seeks to compile existing knowledge on heavy metals in RCP, hypothesize the potential impacts of enforced carbonation, and propose a preliminary framework for evaluating environmental quality of both RCP

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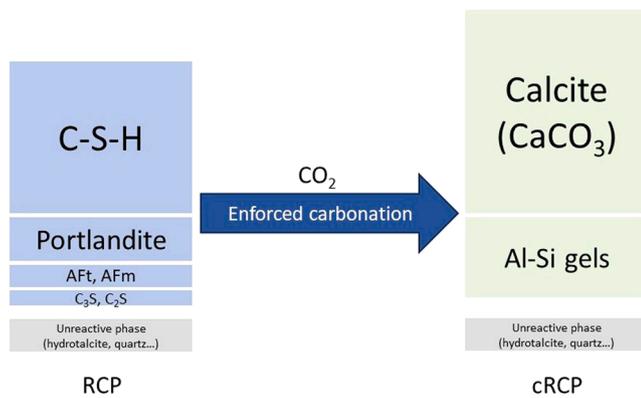


Fig. 1. Change in phase assemblage from RCP to cRCP after enforced carbonation, adopted from the literature (Fang et al., 2021; Zajac et al., 2021b, 2020c).

and the enforced carbonation process.

## 2. Objectives of the review

This review aims to critically examine the current state of knowledge on the immobilization of heavy metals in cement, with a focus on the potential of influence of enforced carbonation to heavy metal behavior in RCP. The review has three main objectives:

- To summarize existing knowledge on the immobilization behaviors of heavy metals in cement systems.
- To consolidate the characteristics of RCP, with a focus on its heavy metal content and potential as SCM within a circular economy context.
- To identify gaps in the literature, concerning the impact of the carbonation on heavy metal phase dynamics and to propose a framework for evaluating immobilization mechanisms in cRCP.

By addressing these gaps, the review establishes a conceptual framework in which enforced carbonation may alter the speciation and retention of heavy metals compared to conventional cement systems. This perspective will contextualize a novel direction for future research, linking carbon capture, circular economy and environmental safety within cement and waste valorization fields.

## 3. Background and overview of heavy metals in cement

Heavy metals are an integral yet concerning component of cement due to their potential environmental and health impact from their leaching. The primary source of heavy metals in cement is raw materials such as limestone, clay, marl, volcanic ash, all of which naturally contain trace amounts of metals like chromium, nickel, and lead (Achternbosch et al., 2003). The use of alternative fuels derived from waste materials such as old tires and municipal solid waste during clinker production via co-incineration is also one of the sources of heavy metal contents in the final cement product (Chatterjee and Sui, 2019; Chen et al., 2009). For example, in Germany, around 69 % of the energy demand of the German cement industry was already substituted with alternative fuels in 2020 (VDZ, 2021). Median contents of heavy metals in the input materials and final products in cement production are listed in Table A1 in Appendix.

Since 1995, the VDZ (Verein Deutscher Zementwerke) has regularly monitored the content of trace elements in standard cements produced in Germany, showing that there has been no significant change over the past 25 years (VDZ, 2021, 1998). Table A2 in Appendix summarizes the heavy metal contents in German standard cements. Despite the increased use of alternative fuels during this period, the contamination of clinker products may not be prominent even for non- or low-volatile

elements such as arsenic, vanadium, lead, and cadmium (VDZ, 2021). In contrast, the incorporation of industrial by-products such as fly ash as an SCM may introduce significant quantities of heavy metals into concrete (Table A3 in Appendix). While the utilization of waste-derived materials offer an appropriate socio-economic solution to mitigate the current challenges in the cement industry, it raises concerns regarding increased heavy metal incorporation in the final concrete products (Achternbosch et al., 2005).

However, total heavy metal content alone is not a reliable indicator of leaching behavior or environmental compatibility of the materials (Van Der Sloot et al., 2011). Thus, leaching tests are employed to estimate the leaching potential of materials by simulating possible weathering scenarios (Malviya and Chaudhary, 2006). Globally, there is a wide diversity of leaching test methods, with each country or region often using its own protocols based on local regulations. For example, in China, GB/T 30810-2014 standard specifies the batch test method to evaluate the heavy metal leaching from cement mortar (GB/T 30810-2014, 2014). In the US, The EPA LEAF Methods 1313-1316 provide protocols for assessing leaching from a wide range of solid materials, however, not federally mandated as standards (U.S. Environmental Protection Agency, 2019). In Europe, efforts to standardize the leaching testing methods began with the Construction Products Directive (CPD), which in 2005 mandated the development of harmonized test methods to assess the environmental performance of construction products. In response, the European Committee for Standardization (CEN) introduced the CEN/TS 16637 series, which has now fully replaced the earlier technical specifications; EN 16637-1 for a general guidance, EN 16637-2 for Dynamic Surface Leaching Test (DSL) for monolithic materials, and EN 16637-3 for percolation test for granular materials (Held et al., 2020). The selection of the appropriate test methods specified in EN 16637-1 is illustrated in Fig. A1. It is important to note that the CPD aimed to harmonize test methods and conformity assessments, but not the regulatory limit values themselves. As a result, member states and private and public sectors are free to define their own environmental requirements. Currently, only a few countries—most notably the Netherlands and Germany—have established maximum leaching limit values for construction products (Overmann et al., 2021). These are applied to results obtained using the DSLT test method in accordance with EN 16637-2 and are summarized as an example in Table A4 in the Appendix (DIN EN 16637-2, 2024). However, the limit values differ significantly between these two countries, highlighting the absence of a unified regulatory protocol. As a result, questions such as “how much heavy metal is too much?” cannot be easily answered in a universal sense, since the definition of acceptable leaching depends entirely on the applicable national regulations and specific leaching methods employed.

Apart from the environmental concern, heavy metals are known to affect the technical properties of cement, such as setting time and compressive strength (Asavapisit et al., 1997; Fernández Olmo et al., 2001; Kitamura et al., 2002; Olmo et al., 2003; Stephan et al., 1999). Several studies have reported that low concentrations of certain heavy metals may enhance mechanical performance. For instance, small amounts of  $Zn^{2+}$  and  $Pb^{2+}$  have shown to improve mechanical properties of cement-based materials (Boardman, 1999; Li et al., 2019). Similarly, CuO has been found to promote  $C_3S$  hydration and ettringite formation and enhance the compressive strength at low dosages (Chen et al., 2007; Ma et al., 2010; Najimi et al., 2011; Tashiro et al., 1979). However, when the concentration of heavy metals exceeds optimal levels, their influence becomes increasingly detrimental to cement performance. Gineys et al. (2010) proposed that the 1 – 3 wt % addition of heavy metals modified the early-age compressive strength due to the delay in alite ( $C_3S$ ) hydration. Similarly, Lu et al. (2017) observed a retardation of  $C_3S$  hydration with the addition of 3 wt %  $Cr^{3+}$ . Zha et al. (2019) also found a detrimental effect on strength development from  $Cr^{3+}$  on cement/fly ash system. Xu et al. (2024) found that  $C_3S$  hydration is strongly retarded by up to 0.15 wt % of the  $Zn^{2+}$  addition, possibly due

to chemisorption of  $Zn^{2+}$  on the  $C_3S$  surface and lowered pH level. The retardation of setting from a high heavy metal concentration has long been attributed to the formation of gelatinous hydroxide surface layer on the cement grains (Thomas et al., 1981). However, Weeks et al. (2008) later suggested the cause of the retardation involves the conversion of metal hydroxides to metal hydroxy-species.

#### 4. Heavy metal content in recycled concrete

CDW primarily consists of concrete, along with bricks, metals, and other materials such as gypsum, plastics, and wood (Bianchini et al., 2005; Nawaz et al., 2023). Recycled concrete is separated from other components of CDW and further processed through smart separation, reaching End-of-Waste status (Skocek et al., 2024). This process yields RCP, a fine fraction of recycled concrete separated from recycled concrete aggregates (RCA) and recycled sand (RS), and RCP is mainly composed of well-hydrated cement (Schoon et al., 2015; Zajac et al., 2021b). Assuming the typical life span of concrete structures is around 70 years, a large number of constructions during the economic growth and the industrial revolution of the 1950s are reaching their EoL in recent years (Pacheco-Torgal et al., 2013). Therefore, the properties, contamination level of the concrete, and construction practices from this era, as well as the effectiveness of the separation process, are particularly relevant for assessing the environmental quality of RCP.

The degree of contamination in CDW is primarily determined by its typology, construction procedures, raw materials used, demolition techniques, and contamination during its service life (Llatas, 2011; Pacheco-Torgal et al., 2013). Given that the heavy metal content in cement has remained constant, as discussed in Section 3, the total heavy metal concentration in RCP may be more influenced by SCMs used in original constructions and pollutants involved in the construction and demolition process. Townsend et al. (2004) explained that heavy metals in CDW can also result from heavy metal-containing debris, such as lead-based paint chips or mercury-containing fluorescent lamps. Yu et al. (2018) discovered that wall insulation and foamed plastic in CDW contain high concentrations of arsenic and cadmium. The use of industrial by-products as clinker replacements began to receive significant attention in the second half of the 20th century (Snellings et al., 2012), which may have contributed to the inclusion of more heavy metals in concrete wastes. Another factor is the lack of stringent regulations on heavy metal content in construction materials during this time, allowing for higher concentrations of such metals during this period. However, before the 1970s, there was no statistical data on the heavy metal content in concrete, as the methodologies for accurately assessing and quantifying such contaminants had not been developed yet in Appendix provides data from recent (2004 – 2020) demolition activities, summarizing total heavy metal contents in RCA, RC and CDWs from various sources.

The total heavy metal content of concrete is determined by the cement, concrete additives, aggregates, and admixtures (Achternbosch et al., 2003). Therefore, theoretical heavy metal content in RCP without the other sources of contamination can be estimated using the heavy metal concentrations in cement and an SCM, as outlined in Table A3 in Appendix (Table 1). For this simplified calculation, potential contributions of heavy metals from admixtures, sand, and coarse aggregates are disregarded, as trace element concentrations from those components are generally negligible (Achternbosch et al., 2003). Assuming a realistic scenario where 30 % of the RCP consists of sand (fine aggregate quartz) after selective separation, the remaining 70 % is composed of a hydrated paste made from cement clinker and SCM mixture. Fly ash, the most common and heavy metal-enriched SCM, was used for this calculation (Snellings et al., 2012). Changes in heavy metal concentrations due to leaching and contamination during the service life are also disregarded. Although this calculation is highly simplified and does not account for numerous factors that significantly influence heavy metal contents in RCP, it could serve as a preliminary assessment.

**Table 1**

Theoretical heavy metal contents of RCP estimated using the heavy metal content in standard cements (VDZ, 1998) and fly ash (Moreno et al., 2005).

0.7 [cement (100 – 70 %) + fly ash (type F) (0 – 30 %)] (w/b = 0.5) + 0.3 sand					
cement:FA Heavy metal	100:0 (mg/kg)	90:10	80:20	70:30	avg.
As	3.7	6.3	8.9	11	7.6
Ba	0.1	70	139	208	104
Be	0.7	1.1	1.4	1.8	1.2
Cd	0.2	0.3	0.4	0.5	0.3
Co	4.6	6.0	7.4	8.8	6.7
Cr	22	28	34	39	30
Cu	17	20	22	25	21
Hg	0.03	0.04	0.05	0.05	0.04
Ni	12	16	20	24	18
Pb	9.1	12	16	19	14
Sb	1.6	1.6	1.7	1.7	1.6
Sn	1.9	2.2	2.4	2.6	2.3
V	27	36	46	55	41
Zn	102	100	98	96	99

It is often the case that a single structure can contain varying concrete types, and a recycling procedure simultaneously treats multiple batches of demolition waste from different sites. Consequently, the chemical and mineralogical profile of RCP, not alone heavy metal concentrations, is highly variable (Zajac et al., 2020c). Heavy metal concentrations in twelve different industrial RCPs are listed in Table 2. These RCPs are obtained via the selective separation of demolished concrete materials, and the origin of raw materials and the grinder used vary between batches. Unlike RCA or recycled concrete fine (RCF), RCP can contain half or more cement hydrates, separated from RCA and RS with a yield of 1 – 15 wt % (Skocek et al., 2024). The hydrated CEM I pastes (a) and (b) are synthetic model systems, designed to simulate a theoretical RCP without any SCMs or chemical additives. Comparing the heavy metal content between industrial RCPs (Table), cement (Table A2 in Appendix), and common SCM materials (Table A3 in Appendix), it is notable that the heavy metal concentrations in RCP are often in a similar range, or even lower than the common SCM materials. This range also aligns with the theoretical heavy metal concentrations of RCP calculated in Table 1, indicating that external contamination from construction and separation practices has minimal impact on the heavy metal content. These findings suggest that RCP does not inherently contain higher levels of heavy metals compared to traditional cement. Typically, recycled concrete is categorized as non-hazardous waste compared to other components separated from CDW (Llatas, 2011; Yu et al., 2018). Many studies have further demonstrated through leaching tests that recycled concrete poses minimal environmental concerns (Bestgen et al., 2016; Engelsen et al., 2010; Gao et al., 2015; Kurda et al., 2018; Lu et al., 2019). However, historical and regional usage patterns of construction materials can influence heavy metal profiles, as shown in

#### 5. Immobilization of heavy metals

Immobilization is the process of rendering hazardous substances less mobile by converting them into stable forms. Cement is one of the most adaptable and effective materials available for the immobilization of heavy metals due to its accessibility and cost-effectiveness (Adaska et al., 1998; Glasser, 1994). In cement systems, heavy metals can be well integrated within the matrix via solidification/stabilization (s/s) process (Chen et al., 2023, 2009; Glasser, 1997; Rađenović et al., 2019).

The strongly alkaline environment (pH 12–13) of the cement system significantly reduces metal solubility. Consequently, precipitation is the dominant immobilization mechanism, with metals forming low solubility species such as metal hydroxides, carbonates, sulfates and silicates (Chen et al., 2023; Glasser and Macphree, 1993; Li et al., 2001). For example, Gineys et al. (2010) suggested that the 1 – 3 wt % addition of  $Cd^{2+}$ ,  $Ni^{2+}$ , and  $Zn^{2+}$  primarily results in the formation of metal

**Table 2**

Heavy metal concentrations in two CEM I pastes and eight different batches of industrial RCPs. (CEM I paste (a): 80 % CEM I 42.5 R and 20 % quartz, CEM I paste (b): 100 % CEM I 52.5 R).

Heavy metal	CEM I paste		Industrial RCP												
	(a)	(b)	Batch 1	Batch 2	Batch 3	Batch 4	Batch 5	Batch 6	Batch 7	Batch 8	Batch 9	Batch 10	Batch 11	Batch 12	
	mg/kg														
As	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	8.1	4.8	< 5	< 5
Ba	111	151	328	351	304	372	240	333	376	235	203	176	363	367	
Be	–	–	1.2	1.2	0.5	0.7	1.4	0.8	1.4	0.2	1.0	0.6	1.2	1.8	
Cd	< 0.4	< 0.4	0.7	0.8	< 0.4	< 0.4	0.8	–	–	< 0.3	0.3	0.4	1.5	0.7	
Co	5.7	< 1	8.0	7.5	4.3	8.1	6.4	–	–	–	8.2	6.9	< 2	< 3	
Cr	70	20	75	94	39	76	67	–	–	–	149	164	63	45	
Cu	108	8	48	50	23	36	30	55	34	28	52	84	11	10	
Hg	–	–	–	–	–	–	–	0.01	0.03	0.09	–	–	0.13	0.01	
Mn	236	192	769	822	436	766	389	–	–	464	696	391	1010	434	
Mo	< 2	< 2	< 2	< 2	< 2	< 2	< 2	< 2	< 2	< 2	2.3	4.2	< 2	< 1	
Ni	26	12	28	26	18	28	26	46	30	25	25	55	13	29	
Pb	< 3	< 3	45	45	< 2	44	20	< 5	41	64	76	10	113	< 5	
Sb	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	1.9	3.1	< 5	< 5	
Se	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	1.7	1.1	< 5	< 5	
Te	–	–	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 0.05	< 0.05	< 5	< 5	
Tl	–	–	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	0.2	0.1	< 5	< 5	
V	52	22	54	51	34	54	51	36	53	45	49	43	32	42	
Zn	110	32	202	215	58	147	86	74	144	116	192	103	223	56	

hydroxides. The sequential chemical extraction study from Li et al. (2001) is in agreement this result, where  $\text{Cu}^{2+}$  and  $\text{Zn}^{2+}$  mainly form hydroxides on the surface of C-S-H. Additionally, the low permeability of hardened cement paste further prevents immobilized heavy metals from migrating through the materials (Conner and Hoeffner, 1998). Cement hydrates are complex mineral phases capable of sorbing heavy metal ions or complexes. Sorption-including chemisorption, physisorption, ion-exchange, physical encapsulation, and structural incorporation-is influenced by surface charge, functional groups, and specifically adsorbed ions of the cement hydrates (Cheng and Bishop, 1992). During the formation of cement hydration products, heavy metal ions can adsorb onto surfaces and then incorporate into the crystal lattice by forming solid solutions, thereby modifying crystallinity, particle size, and solubility of the hydrates (Liu et al., 2023).

This chapter summarizes key findings on this topic to outline the overview of current understanding regarding heavy metal immobilization in cement hydrate phases. Since RCP is largely composed of cement hydrates, understanding these mechanisms is essential to anticipate the initial retention state of heavy metals in RCP.

### 5.1. Calcium silicate hydrate (C-S-H)

C-S-H is the major hydration product of Portland cement and is a gel-like mixture of poorly crystallized compounds with different morphologies. C-S-H gel contains the bulk of micro-porosity, and these gel pores contribute to a high specific surface area (200 – 300  $\text{m}^2/\text{g}$ ) and high sorption capacity of the gel (Chen et al., 2009). Hong and Glasser (2002) proposed that C-S-H gel has a high sorption capacity for binding alkaline metals. Generally, C-S-H can chemically incorporate metals into its structure in silicon sites of Si-O chains and calcium sites of interlayers (Gougar et al., 1996; Qiao et al., 2023). In addition to that, C-S-H can also physically adsorb heavy metals on its surface due to its high specific surface area and unbalanced surface charge (Chen et al., 2009; Gougar et al., 1996). Gougar et al. (1996) suggested that  $\text{Ni}^{2+}$  could intermix with the C-S-H phase or substitute  $\text{Ca}^{2+}$  from its surface. Chen et al. (2009) proposed that almost all calcium in natural crystalline phases of C-S-H can be replaced with heavy metal cations, such as  $\text{Co}^{2+}$  or  $\text{Ni}^{2+}$ . In the case of chromium, Ivey et al. (1990) concluded that  $\text{Cr}^{3+}$  can be incorporated into C-S-H, substituting both calcium and silicon. In contrast, Kindness et al. (1994) found no evidence that trivalent or hexavalent chromium is bound in C-S-H. Gineys et al. (2010) found  $\text{Cu}^{2+}$  and  $\text{Pb}^{2+}$  are trapped in the silica tetrahedra site of C-S-H, while other

researchers suggested otherwise, stating that  $\text{Pb}^{2+}$  is precipitated as an insoluble hydroxide compound (Gougar et al., 1996; Halim et al., 2004; Li et al., 2001). The Richardson-Groves model described that if the amount of present heavy metals during  $\text{C}_3\text{S}$  hydration is sufficient, co-precipitation of heavy metals and calcium can occur, and then resulting heavy metal hydroxides could substitute for  $\text{Ca}(\text{OH})_2$  in C-S-H gel (Richardson, 2000, 1999; Richardson and Groves, 1993; Richardson et al., 2002). Chen (2003) explained that the incorporation of heavy metal cations in C-S-H gel is similar to that seen in glass, where heavy metals behave as network modifiers or network intermediates. C-S-H was also found to physically encapsulate heavy metal compounds such as nickel, cadmium hydroxides and mercury oxide (Cartledge et al., 1990).

### 5.2. Aluminate phases

Ettringite ( $3\text{CaO}\cdot\text{Al}_2\text{O}_3\cdot3\text{CaSO}_4\cdot32\text{H}_2\text{O}$ ) forms in the early stage of hydration of Portland cement. Ettringite and other AFt phases form hexagonal prismatic crystals which consist of  $\{\text{Ca}_6[\text{Al}(\text{OH})_6]_2\cdot24\text{H}_2\text{O}\}^{6+}$  columns and  $\{(\text{SO}_4)_3\cdot2\text{H}_2\text{O}\}^{6-}$  channel components (Fig. 2b) (Gougar et al., 1996; Wang and Wang, 2022). Gougar et al. (1996) described the structure of ettringite as a very ‘forgiving’ one, allowing chemical substitution at calcium, aluminum, and sulfate sites for heavy metals. Ettringite minerals undergo compositional change at  $\text{Ca}^{2+}$ ,  $\text{Al}^{3+}$ ,  $\text{SO}_4^{2-}$  and  $\text{OH}^-$  sites on their structure. Half of the sulfate contents can be substituted by anions, and charge balance is accomplished via substitution, while sacrificing stoichiometry (Gougar et al., 1996). Ions which can substitute  $\text{Ca}^{2+}$ ,  $\text{Al}^{3+}$  and  $\text{SO}_4^{2-}$  sites in ettringite phases are listed in Table A6 in Appendix. While the substitution of  $\text{Ca}^{2+}$  with various divalent heavy metal ions is possible in AFt, the contribution of AFt in a practical Portland cement immobilization system is believed to be weaker than C-S-H (Geysen et al., 2004).

AFm phases are chemically complex, where  $\text{Ca}_6(\text{Al}(\text{OH})_6)\cdot24\text{H}_2\text{O}$  are in lines with interlayer spaces in between and  $\text{Al}(\text{OH})_6$  are octahedrally bonded with three  $\text{Ca}^{2+}$  ions (Fig. 2c) (Chen et al., 2009). The layered structure of AFm can incorporate compounds such as carbonate, sulfate, hydroxide and silicate (Gougar et al., 1996). The chemistry of AFm is therefore dependent on the activities of interlayer-substituent anions (Aimoz et al., 2012). Several studies have demonstrated that the substitution of sulfate in ettringite is an important binding mechanism for oxyanions such as  $\text{CrO}_4^{2-}$ ,  $\text{AsO}_4^{2-}$ , and  $\text{CrO}_4^{2-}$ , and  $\text{CrO}_4^{2-}$  can be incorporated in AFm and AFt as monochromate and  $\text{CrO}_4$ -ettringite

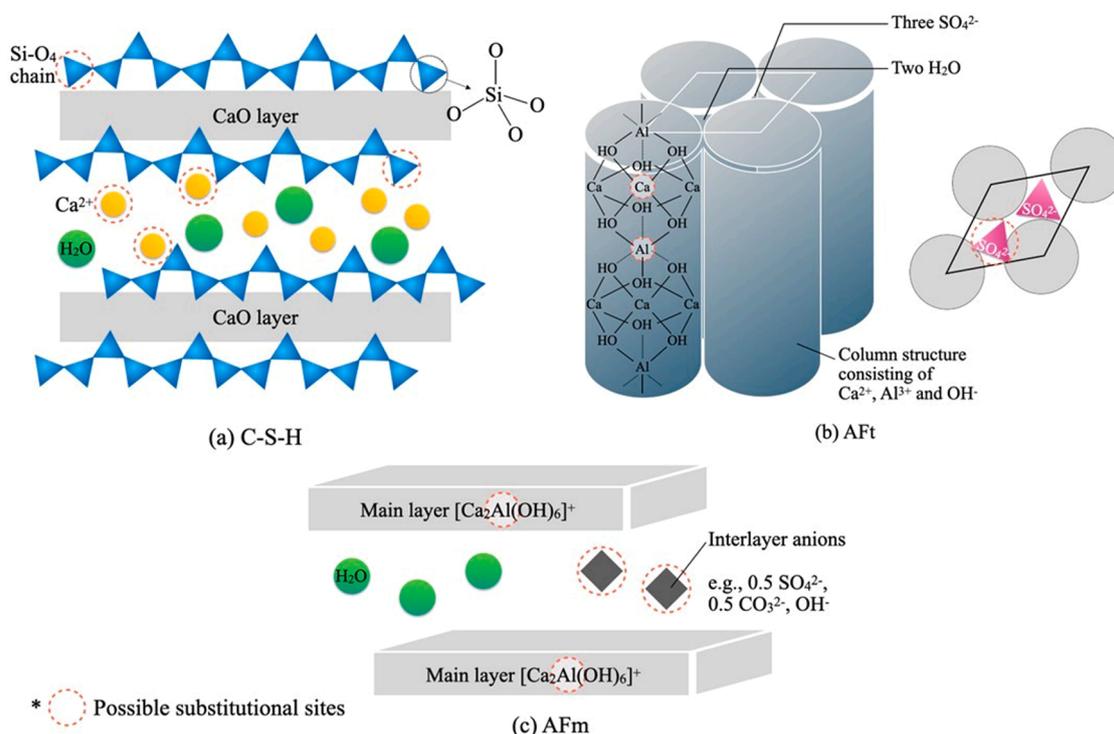


Fig. 2. Possible substitutional sites for heavy metals in the structures of (a) C-S-H, (b) AFt and (c) AFm according to the possible binding mechanisms in the reference (Chen et al., 2009; Gougar et al., 1996; Wang and Wang, 2022).

(Leisinger et al., 2012, 2010).

Although general immobilization mechanisms of heavy metals in cement-based s/s systems have been studied for many years, predicting the behavior of heavy metals in different systems remains challenging. While the role of cement hydrates in heavy metal immobilization has become clearer over time, the numerous potential binding sites and the typically low concentrations of heavy metals make it difficult to determine precise mechanisms in each system.

Macphee and Glasser (1993) provided an early foundation by categorizing the solubility behavior of inorganic metal ions under highly alkaline conditions typical in cement systems (pH 12 – 13). Their classification grouped ions into three categories; soluble ions (e.g. Na, K), irreversible precipitates (e.g. Mn, Ni, Cu), and amphoteric ions which form soluble complexes (e.g. Pb, Cr(III)). According to this model, hydroxide precipitation may be dominant retention mechanism for precipitation-type metals, and amphoteric or soluble ions are more likely to rely on sorption onto cement hydrates. However, experimental findings reported in the literature often deviate from this theoretical categorization. In practice, heavy metal retention mechanisms vary depending on the specific cement system, the presence of supplementary phases, and other environmental variables. To illustrate this complexity, Table 3 summarizes the wide range of immobilization mechanisms reported for  $\text{Cu}^{2+}$ ,  $\text{Mn}^{2+}$  and  $\text{Pb}^{2+}$  across various cement systems, demonstrating that multiple mechanisms can occur conjointly or simultaneously.

## 6. Effect of enforced carbonation on heavy metals

The understanding of heavy metal behaviors in cementitious materials has evolved significantly, with considerable focus on their leaching potential. However, the impact of enforced carbonation on the fate of heavy metals remains unexplored. Many studies have investigated the effects of natural carbonation on heavy metals in cement regarding the correlation between carbonation and heavy metal fixation, and in terms of the durability aspect of the concrete product (Jianguo et al., 2009; Ni

et al., 2017; Valls and Vázquez, 2001; Van Gerven et al., 2004; Zhang et al., 2022). The influence of carbonation on heavy metals mainly results from the changes in the redox potential and pH drop from the carbonation of portlandite (Hobson et al., 2017; Johannesson and Utgenannt, 2001). Several authors reported that the heavy metal retention capacity of hydrated CEM I and anhydrous  $\text{C}_3\text{S}$  has improved after accelerated carbonation (Jianguo et al., 2009; Patterson et al., 1977; You et al., 2011). Greater sorption capacity of decalcified C-S-H gel and calcite, precipitation of other carbonate species, and reduced permeability after carbonation improve the heavy metal immobilization capacity of cement (Chen et al., 2009; Wang and Wang, 2022). Lange et al. (1996) found that lead and zinc are immobilized in the silica-rich area of the carbonated cement as Si-O-M, whereas nickel and chromium appeared to be in the calcite-filled porosity. Zha et al. (2019) found that carbonation increased the leaching of copper and lead from cement-solidified fly ash. Zhu et al. (2024) suggested that 10 – 12 % of accelerated carbonation during the curing of cement paste resulted in a 98 % reduction in leaching concentration for  $\text{Pb}^{2+}$  and 50 % reduction for  $\text{Mn}^{2+}$ . However, the process had an adverse effect on soluble ions such as Cr(VI), due to the reduction in available  $\text{Ca}^{2+}$  ions.

Enforced carbonation, by contrast, is a valorization process intentionally designed to mimic the fundamental chemical reactions of natural carbonation within an engineered setting. These reactions primarily involve  $\text{CO}_2$  interacting with calcium-bearing phases, resulting in the formation of calcium carbonate and aluminosilicate gel, which transforms RCP into cRCP. A comparison and definition of enforced carbonation and accelerated carbonation is summarized in Table A7 in Appendix. Therefore, insights from studies on natural (accelerated) carbonation serve as an initial reference point for understanding heavy metal behavior during the enforced carbonation of RCP into cRCP. Although it shares the same fundamental chemistry, enforced carbonation involves treating finely ground materials with high concentrations of  $\text{CO}_2$  in the presence of  $\text{H}_2\text{O}$ . Depending on the engineering of the process, enforced carbonation can be conducted in two pathways: moist (semi-dry) carbonation with  $\text{H}_2\text{O}$  vapor and wet (aqueous) carbonation

**Table 3**  
Immobilization mechanisms of heavy metals (Cu<sup>2+</sup>, Mn<sup>2+</sup>, Pb<sup>2+</sup>) proposed in the literature.

Heavy metal	Suggested mechanism	Reference	System
Cu <sup>2+</sup>	Hydroxide precipitation	Hashem et al. (2011)	Pure C <sub>3</sub> S with 1 and 3 wt % Cu <sup>2+</sup>
	Substitution of interlayer Ca <sup>2+</sup> and partial substitution of intralayer Ca <sup>2+</sup> in the form of Si-O-Cu	Liu et al. (2024)	Pure C-S-H with 7.83 mM Cu <sup>2+</sup>
Mn <sup>2+</sup>	Surface adsorption, trapped in the silicate tetrahedra of C-S-H	Gineys et al. (2010)	Standard CEM I mortar with 0.18 mol/kg Cu <sup>2+</sup>
	Hydroxide precipitation	Wang and Wang (2022)	Pure anhydrous C <sub>3</sub> S, C <sub>3</sub> A, C <sub>4</sub> AF and C-S-H with 2 – 15 wt % Mn <sup>2+</sup>
Pb <sup>2+</sup>	Related to the formation of AFt and AFm phases	Qian et al. (2006)	Composite cement with CEM I, MSWI fly ash and sludge
	Precipitation of low solubility hydroxide compounds	Li et al. (2001)	Composite cements with CEM I, fly ash and copper sludge
		Halim et al. (2004)	CEM I with 2.3 wt % Pb <sup>2+</sup>
	Trapped in the silicate tetrahedra of C-S-H, forming the end group Si-O-Pb	Gineys et al. (2010)	Standard CEM I mortar with 0.18 mol/kg Pb <sup>2+</sup>
		Wang and Wang (2022)	Pure anhydrous C <sub>3</sub> S, C <sub>3</sub> A, C <sub>4</sub> AF and C-S-H with 2 – 15 wt % Mn <sup>2+</sup>
		Moulin et al. (2000)	Pure pre-hydrated and unhydrated C <sub>3</sub> S, C <sub>3</sub> A
	Ca <sup>2+</sup> ion exchange on the C-S-H surface	Vollpracht and Brameshuber (2016)	CEM I pastes
		Qiao et al. (2023)	Modelling with molecular dynamics simulation
Structural substitution in AFt and AFm	Komarneni et al. (1988)	Natural tobermorite, wollastonite and synthetic xonolite with 1000 mg/L Pb <sup>2+</sup>	
	Qian et al. (2006)	Composite cement with CEM I, MSWI fly ash and sludge	
Chemisorption with monomolecular surface adsorption on ettringite	Fan et al. (2023)	Synthetic ettringite with 2 wt % Pb <sup>2+</sup>	
	Formation of leadhillite and C-Pb-S-H	Lee (2007)	90 % CEM I + 10 % lead hydroxide compounds

in aqueous suspensions (Snellings and Matschei, 2025; Zajac et al., 2022, 2021b). Since carbonation is always mediated by H<sub>2</sub>O in all these scenarios, the wet carbonation is particularly suited for investigating the carbonation mechanism, as monitoring solution evolution provides a direct overview of the mechanisms. Additionally, the high w/b ratio of the wet carbonation increases the mobility of heavy metals during the process, thereby significantly influencing their behavior.

### 6.1. Enforced carbonation of RCP

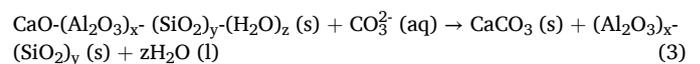
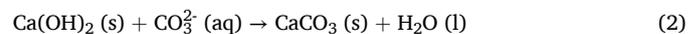
Enforced carbonation falls under the broader concept of CO<sub>2</sub> mineralization which refers to the reaction of metal oxides (MO), such as CaO or MgO, react with CO<sub>2</sub> to form stable, inorganic metal carbonates (eq (1)) (Huijgen, 2007; Jang et al., 2016). The reaction can encompass other species such as alite and belite, initiated with the decalcification of respective minerals, with subsequent carbonation (Zajac et al., 2023a). The thermodynamic stability of the resulting carbonate minerals grants

a permanent CO<sub>2</sub> storage solution (Ben Ghacham et al., 2017; Bobicki et al., 2012, 2012; Chen et al., 2006; Ho et al., 2019a, 2019b).



RCP is one of the materials which can be utilized for this process (Ho et al., 2021; Skocek et al., 2020). Enforced carbonation of RCP can be achieved rapidly at ambient temperature and atmospheric pressure, yielding calcite and a pozzolanic amorphous alumina-silica gels (Al-Si gels) (Shi et al., 2016; Zajac et al., 2020c). The enforced carbonation setup in a laboratory is shown in Fig. A2 in Appendix. In this process, RCP is introduced into a CO<sub>2</sub>-rich solution at ambient temperature and pressure, with CO<sub>2</sub> gas continuously supplied directly into the solution. A high w/b ratio implemented in the process facilitates faster solvation of CO<sub>2</sub>, leading to faster carbonation. When RCP is introduced to the solution in equilibrium with CO<sub>2</sub>-containing gas, the pH level spikes instantaneously due to the rapid dissolution of cement hydrates. It then decreases as carbonation takes effect and carbonation products begin to precipitate. In the carbonation experiment with synthetic RCP made from CEM I 42.5 in 0.1 M NaOH solution, the pH level after the completion of carbonation at equilibrium is about 7 – 7.5 (Zajac et al., 2020c, 2020a).

The reaction kinetics start with portlandite reacting with dissolved CO<sub>2</sub> until it is depleted. Subsequently, other hydration products such as C-S-H, AFt and AFm begin to decalcify (Zajac et al., 2020b). The two main reactions involved in enforced carbonation are presented in Eq. (2) and Eq. (3) (Zajac et al., 2021b). Phases such as ettringite, AFm, monocarbonate and untreated cement components undergo carbonation in a similar manner. The reaction results in the formation of calcite, which precipitates primarily in the voids previously occupied by the solution and on the surface of the cement paste grains. Concurrently, Al-Si gels form in spaces originally occupied by alumina- and silica-bearing hydrates (Zajac et al., 2020c). Aluminum from other carbonatable species, such as ettringite, is also incorporated within Al-Si gels.



While the principle of enforced carbonation is similar to that of natural carbonation, the kinetics differ significantly (Zajac et al., 2020b). Natural carbonation is a slow process spanning several years, where the kinetics are governed by the diffusion of CO<sub>2</sub> gas through the pore network (Ashraf, 2016; Fernández Bertos et al., 2004). On the other hand, wet carbonation involves finely ground powders suspended in a CO<sub>2</sub>-rich solution, resulting in close to full carbonation in less than 2 h (Mao et al., 2023; Zajac et al., 2023a).

Despite multiple commercial decarbonization technologies in the construction sector are currently under development and claim large potential to reduce CO<sub>2</sub> emissions, a recent study by Driver et al. (2024) revealed that enforced carbonation of RCP as the most cost-effective solution. Producing cement with cRCP is the only technology in the study whose cost to reduce CO<sub>2</sub> emission is lower than that of carbon capture and storage (CCS). Notably, the enforced carbonation of RCP was found to be over 20 times more cost-effective than the carbonation of RCA. In this report, RCP represents the largest share of carbonatable solid feedstock-estimated at 0.74 Gt/y from concrete and 0.65 Gt/y from mortar-out of total material stream of 3.93 Gt/y. Given the significant increase in cement production over the past five decades, RCP availability is expected to grow substantially in the coming year. Alongside advances in separation technologies, this trend further enhances the future potential of RCP carbonation technology as a scalable, economically feasible solution (International Energy Agency (IEA), 2018). The first industrial-scale trials are already underway, and the enforced carbonation reactor is undergoing testing in Poland (Fig. A3) (Lloyd-Perks, 2024).



on their surface (Fig. 3c).

### 6.2.1. Heavy metal sorption and coprecipitation on carbonation products

Heavy metals released to the solution via the suggested mechanisms above can be re-immobilized in cRCP. Final cRCP products can include other non-carbonatable phases such as unreacted slag, hydrotalcite and quartz, depending on their original mineralogical compositions (Zajac et al., 2021a, 2020b). However, in this section, the sorption properties of two main carbonation products, calcite and Al-Si gels, will be discussed. This is due to that the inert phases in RCP pastes can vary depending on the original composition of RCP, whereas calcite and Al-Si gels are the universal carbonation products from common cement hydrates.

**6.2.1.1. Calcite.** Calcite is one of three polymorphs of  $\text{CaCO}_3$  and is the most stable under ambient conditions (Plummer and Busenberg, 1982; Zhou et al., 2010). While all three polymorphs, including aragonite and vaterite can be formed during natural carbonation, only calcite is formed from enforced wet carbonation (Zajac et al., 2020c, 2020b).

Calcite is known to chemisorb divalent metal cations ( $\text{M}^{2+}$ ) on its surface by ion-exchange with  $\text{Ca}^{2+}$  (García-Sánchez and Álvarez-Ayuso, 2002; McBride, 1979; Sposito, 1987; Temmam et al., 2000). Heavy metal uptake begins with rapid chemisorption, followed by slower precipitation via the formation of  $\text{MCO}_3(\text{s})$ - $\text{CaCO}_3(\text{s})$  solid solutions (Fuller and Davis, 1987; McBride, 1979; Wersin et al., 1989). Zachara et al. (1991) explained the surface-exchange reaction model for the divalent metal sorption behaviors of calcite in detail (eq (4)). Their work suggested that divalent metal sorption on calcite is dependent on the aqueous  $\text{Ca}^{2+}$  concentration, proving the model's eligibility. The selectivity of metals in the competitive sorption is explained by similarity in ionic radii ( $r_x$ ) of metal ions in comparison to  $\text{Ca}^{2+}$  (Temmam et al., 2000). Metals with similar or smaller  $r_x$  than  $\text{Ca}^{2+}$  form  $\text{MCO}_3$  surface structure more easily, exhibiting stronger sorption and metal-specific selectivity (Kim et al., 2023; Zachara et al., 1991).



The study of  $\text{Zn}^{2+}$  adsorption on calcite proposed that sites available for  $\text{Zn}^{2+}$  sorption were less than 10 % of  $\text{Ca}^{2+}$  sites on the calcite surface, and the surface exchange reaction did not influence the solubility of calcite (Zachara et al., 1991, 1988). The sorption amount of  $\text{Zn}^{2+}$  increased in higher pH, however, they concluded that this result was from lowered  $\text{Ca}^{2+}$  activities, not from its surface charge. In the study of  $\text{Cd}^{2+}$  adsorption on calcite surface of Davis et al. (1987), the sorbed  $\text{Cd}^{2+}$  exchanged  $\text{Ca}^{2+}$  in the hydrated layer of calcite within 24 h of reaction. The following desorption experiment showed a slower rate, suggesting that the model for divalent metal sorption on calcite is based on a kinetic model, rather than on the assumption of local equilibrium.

Since enforced carbonation is a dissolution-precipitation process, heavy metals may become incorporated into the calcite structure during crystal growth via coprecipitation, as well as adsorbing onto the surfaces of pre-existing calcite formed during enforced carbonation (Gebrehiwet et al., 2012; Hunter et al., 2021; Li et al., 2020). Thermodynamics of heavy metal partitioning in solid solution coprecipitation are explained with solid solution-aqueous solution (SS-AS) theory, where two solutes crystallize simultaneously when their crystal structures are compatible (Prieto, 2009). Metals whose carbonates share the same crystal structure as calcite are more likely to incorporate into the calcite lattice, forming solid solutions (Davis et al., 1987; Drake et al., 2018; Kim et al., 2023). In contrast, metals whose carbonates have incompatible structures, such as  $\text{Ba}^{2+}$ ,  $\text{Ni}^{2+}$  and  $\text{Pb}^{2+}$ , are more likely to interact with calcite surfaces through sorption at the later stage of enforced carbonation, rather than being incorporated into the lattice.

Hunter et al. (2021) demonstrated that heavy metal coprecipitation from wastewater into  $\text{CaCO}_3$  via direct gaseous carbonation was significantly greater than predicted by thermodynamic SS-AS model. In a comparative study of two carbonation methods ( $\text{CO}_2$  bubbling and

$\text{Na}_2\text{CO}_3$  addition),  $\text{CaCO}_3$  precipitated effectively removed heavy metals from solution, with  $\text{CO}_2$  bubbling showing greater efficiency in coprecipitating  $\text{Cu}^{2+}$ ,  $\text{Pb}^{2+}$ , and  $\text{Zn}^{2+}$  (Wang et al., 2016).

**6.2.1.2. Al-Si gels.** Surface characterization of Al-Si gels is not yet well theorized, primarily due to their amorphous nature and in-situ formation during the carbonation. The Al-Si gels have a porous structure with a high surface area, thus contributing to high sorption capacity of cRCP (Mao et al., 2024). Several studies have demonstrated that the BET ( $\text{N}_2$ ) surface area of RCP has increased up to 10 times after enforced carbonation (Shen et al., 2022; Teune et al., 2023). Mao et al. (2023) reported that the silica gel component in cRCP reached up to  $606 \text{ m}^2/\text{g}$  of surface area. In this review, amorphous silica gel is referenced to predict the sorption characterisation of the Al-Si gels formed during carbonation based on three points: a) Al-Si gels are composed of a network of  $\text{SiO}_4$  tetrahedra (Sevelsted and Skibsted, 2015), which shares structural characteristics with amorphous silica; b) Both types have surfaces terminated with hydroxyl (OH) group (Mao et al., 2024); c) Al/Si ratio of carbonated products from the reference RCPs with CEM I 42.5R (Zajac et al., 2020c) is only 0.06, allowing us to disregard the consideration of alumina for simplicity.

The surface of silicon atoms has a complete tetrahedral configuration and in an aqueous solution, their free valence becomes saturate with hydroxyl groups, forming silanol groups (Si-OH). The sorption of metal ions by silica gels is attributed to surface complexation formation with silanol groups, expressed as eq (5) (Schindler et al., 1976). The reaction involves a cation-exchange mechanism, where surface silanol groups release protons ( $\text{H}^+$ ), uptaking metal cations as a weak acid cation exchanger. The affinity sequence of divalent metal sorption, reported by Tran et al. (1999) is  $\text{Pb}^{2+} > \text{Cu}^{2+} > \text{Ni}^{2+} > \text{Zn}^{2+} > \text{Cd}^{2+}$ .



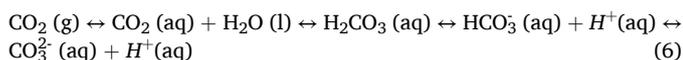
For divalent metals, two moles of  $\text{H}^+$  ions are released per mole of  $\text{M}^{2+}$  ions sorbed on the surface. Given the nature of the acid cation exchange reaction, the surface adsorption is strongly affected by its pH level, as it alters the acidity of the surface silanol groups (Mustafa et al., 2003). Maximum sorption capacity will then be affected by pH level and specific surface area. You et al. (2011) found that the cation exchange capacity (CEC) of carbonated hardened cement paste increased linearly with an increase of the carbonation percentage. This phenomenon was explained with the formation of silica gels with a structural imperfection caused by decalcification. Ouyang et al. (2024) reported that binding energy has shifted following sorption of  $\text{Cd}^{2+}$ ,  $\text{Co}^{2+}$ , and  $\text{Ni}^{2+}$  onto cRCP, demonstrating the reactivity of deprotonated surface hydroxyls-originating from silica gels-toward divalent metal ions.

In the cRCP system, both calcite and Al-Si gels are expected to exhibit high sorption capacity. Several authors concluded that the heavy metal retention ability of cement improves as a result of the carbonation, suggesting that carbonated cement phases can better adsorb heavy metal ions (Lange et al., 1996; You et al., 2011). It is worth noting that under enforced wet carbonation, the surface characteristics of the cRCP-which ultimately determine its retention capacity-can vary depending on factors such as the mineralogical compositions of the starting material, pH level, carbonation duration, and conditions like temperature,  $\text{CO}_2$  pressure, and the solution system (Ashraf and Olek, 2018; Zajac et al., 2024).

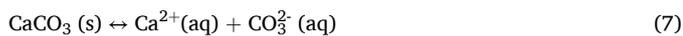
### 6.2.2. Metal carbonate precipitation

The most prominent factor in the solution dynamics during the enforced carbonation is the high concentration of carbonate ion,  $[\text{CO}_3^{2-}]$  and the resulting pH level. In enforced carbonation,  $\text{CO}_2$  gas is continuously supplied in solution to ensure full carbonation of calcium-bearing species with calcium content up to 62 wt % for the CEM I 42.5R RCP paste used in the reference (Zajac et al., 2020c) under a stable pH level. The solution composition adjusts to maintain dynamic equilibrium for

the CO<sub>2</sub>-carbonate-bicarbonate system (eq (6)). The principle where calcium species are fully carbonated to form calcium carbonates can be directly applied to predict a similar tendency for divalent heavy metals. Although metal carbonate precipitation during accelerated carbonation has been reported by several authors (Jianguo et al., 2009; Ni et al., 2017), in enforced carbonation, heavy metal species can dissolve far more easily as a free ion and react with dissolved CO<sub>3</sub><sup>2-</sup>. Therefore, carbonate precipitation phenomena during enforced carbonation may be more prominent in comparison to natural or accelerated carbonation.



Carbonate ions then react with calcium to precipitate CaCO<sub>3</sub> until calcium is exhausted, and the carbonate ion concentration adjusts to the level at its new equilibrium. Once all available calcium is consumed, the concentration of remaining calcium, [Ca<sup>2+</sup>] is then determined by its K<sub>sp</sub> value of CaCO<sub>3</sub> (eq (9)). When the reaction quotient  $Q = [\text{Ca}^{2+}][\text{CO}_3^{2-}]$  is much greater than the K<sub>sp</sub> (eq (8)), the solution is supersaturated with respect to CaCO<sub>3</sub>. When the ionic product Q surpasses the equilibrium threshold, the imbalance between Q and K<sub>sp</sub> drives the system to precipitate the solid product, restoring equilibrium by reducing the concentration of ions in solution (Haynes et al., 2016).



$$Q^{\text{CaCO}_3} = [\text{Ca}^{2+}][\text{CO}_3^{2-}] \quad (8)$$

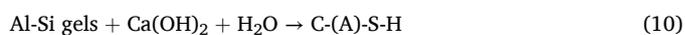
$$K_{\text{sp}}^{\text{CaCO}_3} = 3.36 \times 10^{-9} \quad (9)$$

The nature of the enforced carbonation environment pushes the equilibrium of  $Q \gg K_{\text{sp}}$  throughout the process with high CO<sub>2</sub> concentration, resulting in near full transition of calcium-bearing minerals towards calcite (Zajac et al., 2020b). The system is designed to keep its continuous supply of CO<sub>2</sub> gas, and a similar equilibrium between M<sup>2+</sup>, CO<sub>3</sub><sup>2-</sup> and MCO<sub>3</sub>(s) may happen in the solution. Given that 80 % of the final calcium carbonate content was formed within the first 23 min of carbonation (Zajac et al., 2020c), some metal carbonates may also precipitate in a relatively rapid kinetics at the early stage of enforced carbonation. As predicted at the beginning of this chapter, the solubility of most metal hydroxides formerly present in RCP will increase upon completion of carbonation due to decreased pH level from 12 – 13 to as low as 7 (Feitknecht and Schindler, 1963). With a constant supply of [CO<sub>3</sub><sup>2-</sup>], a certain amount of metal hydroxides could transition to form metal carbonates based on the relative solubility of the given hydroxide and carbonate forms. K<sub>sp</sub> values of heavy metal carbonates and hydroxides at 25 °C are listed in Table A8 in Appendix. CaCO<sub>3</sub> and Ca(OH)<sub>2</sub> are included in the table as a reference. Notably, all listed heavy metals except for nickel have lower K<sub>sp</sub> values than CaCO<sub>3</sub>. By comparing K<sub>sp</sub> values, metal hydroxides such as Cu(OH)<sub>2</sub> and Pb(OH)<sub>2</sub> may precipitate more easily due to their extremely low K<sub>sp</sub> values, assuming the [M<sup>2+</sup>]<sub>threshold</sub> for the hydroxide form of these metals will be in the range of  $\approx 10^{-6}$  in pH 7. In contrast, barium could favor barium carbonate formation, as the solubility of barium hydroxide is sufficiently higher than its carbonate form, similar to calcium. Pourbaix diagrams (Eh-pH diagrams) of heavy metal-CO<sub>2</sub>-H<sub>2</sub>O system are illustrated in Fig. A4. It should be noted that the presence of other ions, such as sulfate (SO<sub>4</sub><sup>2-</sup>), needs to be considered depending on the system of interest. For example, barium may favor the formation of barium sulfate (BaSO<sub>4</sub>), as the solubility of BaSO<sub>4</sub> (K<sub>sp</sub> = 1.1 × 10<sup>-10</sup>) is lower than that of its the carbonate counterpart (Utton et al., 2011).

## 7. Mechanisms of the heavy metal immobilization in composite cement with cRCP

The two main components of cRCP, calcite and Al-Si gels, undergo distinct hydration reaction mechanisms in composite cement. The

calcite component influences hydration similarly to limestone in composite cements, while Al-Si gels act as a pozzolanic SCM (Teune et al., 2023; Zajac et al., 2024). Al-Si gels are proven to react unusually fast compared to traditional SCM materials, forming C-(A)-S-H phase via pozzolanic reaction at an early age (eq (10)) (Mao et al., 2024; Zajac et al., 2024). Given the hydration characteristics of cRCP, the potential immobilization mechanism during enforced carbonation, as described in Section 6.2, suggests two new standpoints on the fate of heavy metals; a) Certain heavy metals may precipitate as metal carbonates or coprecipitate with CaCO<sub>3</sub>. b) Heavy metals adsorbed onto the surface of Al-Si gels will be susceptible to rapid release into the pore solution, possibly during the early hydration phase (< 7 days) (Song et al., 2025; Zajac et al., 2024).



Once cRCP is incorporated with cement clinkers, immobilized heavy metal species again undergo chemical shift in their dynamics. The most prominent change is the pH increase in pore solution. Entering back to the high pH pore solution environment will induce significant changes in heavy metal speciation. In case of heavy metals sorbed onto calcite, given the phenomena where the surface sorption capacity of calcite increases at higher pH (Zachara et al., 1991), release of sorbed metals from calcite may not be as prominent as that of Al-Si gels. The most used metal carbonate in cement production is CaCO<sub>3</sub>, which is proven to have a great filler effect and natural carbonation resistance in composite cements (Fang et al., 2021; Fante et al., 2022; Matschei et al., 2007; Matschei and Glasser, 2010; Voglis et al., 2005). Matschei et al. (2007) suggested that while calcite primarily acts as a filler when used in a large volume, adding up to 5 wt % of calcite can alter the mineralogy of AFm phases. According to the calculated phase assemblages in the literature, at a molar bulk CO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio 0.5 – 1.0, calcite acts as a reactive admixture and is consumed (Matschei et al., 2007). Referencing the properties of CaCO<sub>3</sub> as a clinker substitute can provide insight into how other metal carbonates may behave when introduced to cement clinker. Portland cement incorporating barium carbonate, BaCO<sub>3</sub>, is another example of metal carbonate utilization in cement reported by several authors. With 30 wt % of BaCO<sub>3</sub> replacement in CEM I and 9:1 BFC:CEM I system, the precipitation of barite (BaSO<sub>4</sub>) is observed during hydration (Dumitru et al., 1999; Utton et al., 2011). BaSO<sub>4</sub> being significantly less soluble than the carbonate, when BaCO<sub>3</sub> reacts with Portland cement, all available sulfate is consumed to form BaSO<sub>4</sub> (Utton et al., 2011). The subsequent release of CO<sub>3</sub><sup>2-</sup> ions leads to the formation of monocarboaluminate, in align with the scenario with CaCO<sub>3</sub>. In case of another alkaline earth metal carbonate, strontium carbonate (SrCO<sub>3</sub>), similar phenomena were observed. Walling et al. (2023) proposed that ground granulated blast furnace slag (GGBFS) cement with the addition of 30 wt % of SrCO<sub>3</sub> follows a similar reaction pathway and phase formation to those with CaCO<sub>3</sub>. Minor reactive portion of SrCO<sub>3</sub> leads to the formation of carbonate-AFm phase, and the rest is well-immobilized via physical encapsulation within hydrate phases. No reaction between SrCO<sub>3</sub> and sulfate was observed.

## 8. Challenges and knowledge gaps

### 8.1. Retention mechanisms of heavy metals in cRCP

The effect of carbonation on heavy metal behavior is a well-researched area, particularly in the context of natural carbonation, which occurs gradually through weathering over the service life of concrete structures. A significant body of literature explores how natural carbonation affects the binding and release of heavy metals from cement hydrates. Since enforced carbonation leverages the same underlying chemical reactions, studies on the effect of natural carbonation on heavy metal behaviors are referenced in this review.

However, despite this conceptual link, the mechanistic

interpretation of heavy metal immobilization mechanisms upon carbonation remains inconsistent across studies. In many studies on carbonation of cementitious materials, its effects on heavy metals and the interpretation of experimental results are often vague, particularly regarding the clear distinction and between heavy metal carbonate precipitates and coprecipitation or sorption onto  $\text{CaCO}_3$ . Some studies have described that the retention of heavy metals in  $\text{CaCO}_3$  using terms such as coprecipitation, scavenging, ion-exchange, as explicitly used in the references (Li et al., 2020; Ouyang et al., 2024; Schwartz and Ploethner, 2000; You et al., 2011). In contrast, others have suggested that heavy metals may have precipitated as separate carbonates alongside  $\text{CaCO}_3$  (Jianguo et al., 2009; Ni et al., 2017). To address this gap, future research should focus on detailed mechanistic investigations as well as modelling prediction to identify the contributions of specific mechanisms and critical heavy metal concentrations under enforced carbonation.

Considering the high early-stage reactivity of cRCP, heavy metals initially retained, especially on Al-Si gels, are likely to be released during early hydration and subsequently incorporated into cement hydration products through known immobilization mechanisms, as discussed in Section 5. Thus, one of the critical factors in assessing long-term environmental safety is the quantitative heavy metal retention capacity of cRCP for various heavy metals. A key challenge in evaluating the heavy metal sorption capacity of carbonated materials lies in managing pH and precipitation. Although acidification is widely used to prevent or minimize precipitation during sorption isotherm experiments (Flieger et al., 2020; Ouyang et al., 2024), it greatly risks destabilizing the sorbent materials, distorting both the materials and the results. Main products of enforced carbonation-metal carbonates and Al-Si gels are particularly susceptible to decomposition and highly sensitive to acidic environments (Lu et al., 2018; Mao et al., 2023). Thus, it is plausible that the measured sorption capacity may be underestimated, as the  $\text{CaCO}_3$  matrix is significantly diminished, and the net surface charge of Al-Si gels is reduced due to surface protonation. To accurately assess the heavy metal retention capacity, testing methodologies must be tailored to reflect the chemical stability of carbonation products (e.g. in-situ methodology in equilibrium with  $\text{CO}_2$ ).

Quantification and characterization of respective phases is another critical gap. To validate the hypotheses discussed in this review, accurate characterization between metal carbonates, hydroxides, and sorption is essential. Thermogravimetric and mass spectrometric (TG-MS) and differential thermal analysis (DTA) can provide indirect quantification of carbonate phases (Kemp et al., 2022). However, distinguishing coexisting metal carbonate species with similar thermal properties can be challenging. X-ray diffraction (XRD) can be a useful phase identification method (Ali et al., 2022), but only when crystalline phases are present with above detection limits. Therefore, a multi-analytical approach-combining thermal analysis, XRD, spectroscopy (e.g., FTIR or Raman), and surface characterization techniques such as SEM-EDS or XPS-should be adopted to improve the accuracy and reliability of phase-specific identification and quantification. This integrated strategy is essential for distinguishing between amorphous, crystalline, and surface-bound species, particularly in complex carbonation systems.

## 8.2. Assessment of environmental compatibility

It is important to differentiate the role of natural and enforced carbonation when addressing their impact on leaching of heavy metals. Natural carbonation directly influences the mobility and leachability of heavy metals over time. Accelerated carbonation methods applied to monolithic materials are often used to simulate real-life weathering conditions to evaluate the impact on leaching characteristics as carbonation progresses.

In contrast, enforced carbonation is a treatment applied to fine RCP to produce cRCP, which serves as a novel SCM. The process has no direct influence on the leachability of heavy metals during the service life of

the final concrete product. Rather, it alters the chemical and physical properties of the RCP, thereby influencing how heavy metals in cRCP would interact with cement clinker phases during hydration. Thus, the environmental safety of cRCP should be evaluated within the context of the composite cement system it becomes part of (see Section 7). To date, no study has examined the long-term leaching behavior of heavy metals from cementitious system containing cRCP. Therefore, a combined testing strategy is recommended: monolithic leaching tests on pozzolanic cement mortar or concrete incorporating cRCP, alongside pH-dependence batch leaching tests, to evaluate the heavy metal release over time as well as the leaching characteristics.

## 9. Conclusion and final remarks

With the growing use of alternative and recycled materials in the construction industry, assessing the sustainability of cement has become even more critical. Enforced carbonation of RCP offers a promising strategy to promote both waste recycling and  $\text{CO}_2$  emission reduction, aligning with the principles of the circular economy. This approach leverages the abundant availability of RCP and its significant valorization potential for carbon mineralization. Given that the SCM performance of cRCP in composite cement is already established (Zajac et al., 2024, 2023b), this review focused on evaluating the environmental quality of RCP, particularly its heavy metal content. Both theoretical assessments and actual measurements from industrial RCPs indicate low levels of heavy metals in RCP, ensuring the first-hand material safety of RCP for the enforced carbonation process.

Enforced carbonation fundamentally modifies RCP's phase assemblage and chemical environment, which likely governs heavy metal immobilization through three key mechanisms (Fig. 3); 1) coprecipitation with  $\text{CaCO}_3$ , 2) sorption onto Al-Si gels, and 3) formation of heavy metal carbonate precipitates. Two main products of RCP carbonation-calcite and Al-Si gels-both are expected to demonstrate great heavy metal retention capacity due to their large surface area, and coprecipitation sites on  $\text{CaCO}_3$  via ion-exchange. The rapid precipitation kinetics of calcite in enforced carbonation suggest that similar kinetics may govern the formation of heavy metal carbonates, possibly enhancing the overall heavy metal retention in cRCP. This mechanism also opens possibilities for integrating wastewater treatment within the enforced carbonation process, producing value-added cRCP and removing heavy metals from contaminated water streams.

This study is the first systematic analysis addressing heavy metals in RCP and the effect of enforced carbonation, proposing an evaluation framework and future research directions (Fig. 4). To ensure the safe and widespread use of the material, monitoring and archiving heavy metal contents in RCP prior to enforced carbonation is important. Detailed characterization of heavy metal speciation post-carbonation and assessment of the carbonation solution as well as the investigation on heavy metal retention capacity are critical next steps. Finally, long-term leaching studies on cement composites containing cRCP are essential to evaluate the stability and immobilization of heavy metals in the final product, thereby supporting safe, sustainable material reuse and closing the circular loop.

## CRedit authorship contribution statement

**Kristen Sodam Park:** Writing – review & editing, Writing – original draft, Visualization, Investigation, Formal analysis, Data curation, Conceptualization. **Maciej Zajac:** Writing – review & editing, Supervision, Resources, Project administration, Conceptualization. **Thomas Matschei:** Supervision. **Anya Vollpracht:** Writing – review & editing, Supervision, Investigation, Conceptualization.

## Declaration of competing interest

The authors declare that they have no known competing financial

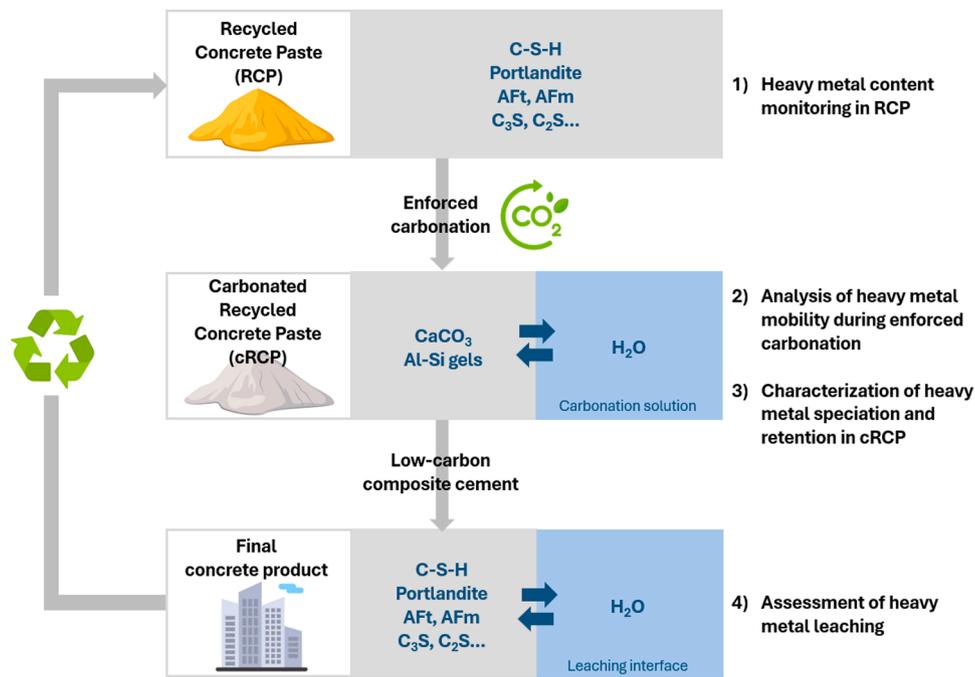


Fig. 4. Schematic representation of the proposed framework linking enforced carbonation with heavy metal monitoring and environmental assessment in circular cement systems.

interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix

Tables A1, A2, A3, A4, A5, A6, A7, A8

Table A1

Median contents of heavy metals in the input materials and final products in the reference (Achtenbosch et al., 2003; Chatterjee and Sui, 2019; VDZ, 1998).

Heavy metal	Intermediate product	Conventional fuels		Alternative (secondary) fuels		Final product	
	Raw meal mg/kg	Hard coal	Oil coke	Used tires	ETP sludge & chemical process residues	Clinker	Portland Cement
As	9	9	0.5	1.6	0.7	9	8
Be	1	1.4	0.03	0.3	-	1.4	1
Cd	0.3	1	1	7	< 1	1	0.6
Co	7	9	2.5	30	1.2	13	11
Cr	30	14	4.3	137	5.1	66	68
Cu	14	18	2.4	68	227	38	38
Hg	0.06	0.3	0.05	0.4	< 0.5	0.1	0.3
Mn	236	58	-	189	49	400	606
Ni	20	23	263	90	13	38	45
Pb	14	27	13	125	3.8	24	27
Sb	0.7	1	0.6	136	-	5	5
Sn	3	4	0.3	15	2.9	13	3
Tl	0.5	1	0.5	10	< 1	0.5	0.6
V	37	39	758	19	4	57	74
Zn	34	63	16	6100	9.6	113	164

Table A2

Heavy metal contents in 418 standard cement samples in 1998 (VDZ, 1998).

Heavy metal	Mean value mg/kg	Range	Number of samples below the detection limit
As	7.0	< 1 - 100	20
Ba	-	< 14,990	-
Be	1.3	< 0.1 - 6	1
Cd	0.4	< 0.1 - 1.5	26

(continued on next page)

**Table A2** (continued)

Heavy metal	Mean value mg/kg	Range	Number of samples below the detection limit
Co	8.7	< 1 - 30	0
Cr	41	< 1 - 200	0
Cu	31	< 1 - 250	0
Hg	0.06	< 0.1 - 0.5	123
Mn	759	< 200 - 5000	0
Mo	–	< 14	–
Ni	23	< 1 - 60	0
Pb	17	< 0.1 - 100	0
Sb	2.9	< 0.1 - 53	61
Se	<sup>a</sup> n.d.	–	415
Sn	3.6	< 1 - 30	29
Te	n.d.	–	415
Tl	0.4	< 0.5 - 5	372
V	50	< 20 - 200	0
Zn	192	< 1 - 1100	0

<sup>a</sup> n.d.: not detectable.**Table A3**

A comparison of average heavy metal concentrations between standard cement and common SCMs.

Heavy metal	Standard cement (VDZ, 1998) mg/kg	Blast-furnace slag (Achterbosch et al., 2003)	Clay (Achterbosch et al., 2003)	Fly Ash (Moreno et al., 2005)	Coal fly ash (Achterbosch et al., 2003)	Limestone (Achterbosch et al., 2003)
As	7.0	0.8	14	55	79	3
Ba	<sup>a</sup> n.d.	–	–	1302	–	–
Be	1.3	–	3	8	15	0.3
Cd	0.4	0.7	0.2	2	2.6	0.2
Co	8.7	4	20	35	74	3
Cr	41	25	85	148	127	14
Cu	31	5.2	43	86	247	11
Hg	0.06	0.6	0.2	0.2	0.3	0.04
Mn	759	3500	600	–	484	500
Ni	23	5	63	96	196	18
Pb	17	6	25	80	257	18
Sb	2.9	2	2	4	14	1
Se	n.d.	1.5	0.5	7	8	0.6
Sn	3.6	5	5	8	10	4
Te	n.d.	–	0.5	–	1.6	0.7
Tl	0.4	0.7	0.5	–	4	0.3
V	50	30	130	228	345	26
Zn	192	38	78	154	504	30

<sup>a</sup> n.d.: not detectable.**Table A4**

Leaching limit values set for construction products in Germany and the Netherlands (Deutsches Institut für Bautechnik (DIBt), 2025; Ministerie van Infrastructuur en Waterstaat, 2008).

Heavy metals	DIBt MVV TB 2025, hardened concrete with sulfate-resistant cement, calcium aluminate sulfate cement, or other industrially manufactured aggregates mg/m <sup>2</sup>	DIBt MVV TB 2025, hardened concrete with silicon-rich fly ash or boiler sand	Dutch Soil Quality Decree 2008
As	18		260
Ba	979	979	1500
Cd	1.7		3.8
Co	11		60
Cr	19	19	120
Cu	30		98
Hg	0.56	0.56	1.4
Mo	196		144
Ni	39		81
Pb	6.7	6.7	400
Sb	28		8.7
Se	17	17	4.8
Sn			50
Tl	1.1	1.1	
V	22	22	320
Zn	336	336	800

**Table A5**  
Heavy metal concentrations in RCAs and CDWs from various sources.

Heavy metal	<sup>a</sup> RCA Shanghai and Sichuan, China (Gao et al., 2015) n = 4 mg/kg	<sup>b</sup> RCA Norway (Engelsen et al., 2010) n = 4	<sup>c</sup> RCA the U.S. (Chen et al., 2013) n = 7	<sup>d</sup> CDW debris Florida, the U.S. (Townsend et al., 2004) n = 5	<sup>e</sup> Demolished concrete West China (Chen and Zhou, 2020) n = 80
As	–	–	5.4	2.7 ± 2.6	78
Ba	–	–	66	–	–
Cd	–	< 0.7	0.2	1.1 ± 3.6	1.7
Co	–	–	2.3	–	–
Cr	70.7 ± 12.8	90	9.6	18 ± 1.6	71
Cu	24.6 ± 2.9	35	11	34 ± 1.9	19
Hg	–	–	–	0.16 ± 3.1	< 1.0
Mn	–	557	–	–	< 0.05
Mo	–	6	0.6	–	–
Ni	21.3 ± 1.8	28	7.0	51 ± 1.7	19
Pb	23.3 ± 3.8	43	4.9	66 ± 2.3	58
Zn	906 ± 538	228	32	200 ± 2.0	67
V	–	77	–	–	–

<sup>a</sup> Mean ± standard deviations.

<sup>b,c,e</sup> Mean.

<sup>d</sup> Arithmetic mean ± standard deviations.

**Table A6**  
Reported ion substitutions in ettringite phases in the literature  
(Chen et al., 2009; Gougar et al., 1996; Leisinger et al., 2012, 2010; Tashiro and Oba, 1979).

Ca <sup>2+</sup> site	Al <sup>3+</sup> site	SO <sub>4</sub> <sup>2-</sup> site
Sr <sup>2+</sup>	Cr <sup>3+</sup>	CO <sub>3</sub> <sup>2-</sup>
Ba <sup>2+</sup>	Si <sup>4+</sup>	Cl <sup>-</sup>
Pb <sup>2+</sup>	Fe <sup>3+</sup>	OH <sup>-</sup>
Cd <sup>2+</sup>	Mn <sup>3+</sup>	CrO <sub>4</sub> <sup>2-</sup>
Co <sup>2+</sup>	Ni <sup>3+</sup>	AsO <sub>4</sub> <sup>2-</sup>
Ni <sup>2+</sup>	Co <sup>3+</sup>	NO <sub>3</sub> <sup>-</sup>
Zn <sup>2+</sup>	Ti <sup>3+</sup>	SO <sub>3</sub> <sup>2-</sup>

**Table A7**  
Comparison of mineral carbonation techniques (Snellings and Matschei, 2025).

	Enforced carbonation	Accelerated carbonation
Objectives	Engineered mineral carbonation reaction for preparation of reactant materials	Assessment of deterioration by exposure to CO <sub>2</sub> , in context of concrete durability or leachability
CO <sub>2</sub> concentration	High (up to 100 %)	Moderate (e.g. 1 – 3 %)
Processing	Wet (aqueous suspension)    Moist (semi-dry)	Moist (semi-dry)
Target materials	Ground materials e.g. Carbonatable binders including recycled concrete paste (RCP), aggregate (RCA), and fines (RCF)	Monolithic materials e.g. Mortar, concrete

**Table A8**  
K<sub>sp</sub> of divalent metal carbonates and hydroxides at 25 °C (Grauer, 1999; Haynes et al., 2016; Patnaik, 2003).

Carbonate compound	K <sub>sp</sub>	Hydroxide compound	K <sub>sp</sub>
BaCO <sub>3</sub>	2.58 × 10 <sup>-9</sup>	Ba(OH) <sub>2</sub> •8H <sub>2</sub> O	2.55 × 10 <sup>-4</sup>
CdCO <sub>3</sub>	1.00 × 10 <sup>-12</sup>	Cd(OH) <sub>2</sub>	7.20 × 10 <sup>-15</sup>
CaCO <sub>3</sub>	3.36 × 10 <sup>-9</sup>	Ca(OH) <sub>2</sub>	5.02 × 10 <sup>-6</sup>
CoCO <sub>3</sub>	7.94 × 10 <sup>-12</sup>	Co(OH) <sub>2</sub>	5.92 × 10 <sup>-15</sup>
CuCO <sub>3</sub>	3.55 × 10 <sup>-12</sup>	Cu(OH) <sub>2</sub>	2.20 × 10 <sup>-20</sup>
MnCO <sub>3</sub>	2.24 × 10 <sup>-11</sup>	Mn(OH) <sub>2</sub>	5.61 × 10 <sup>-12</sup>
NiCO <sub>3</sub>	1.42 × 10 <sup>-7</sup>	Ni(OH) <sub>2</sub>	5.48 × 10 <sup>-16</sup>
PbCO <sub>3</sub>	7.40 × 10 <sup>-14</sup>	Pb(OH) <sub>2</sub>	1.43 × 10 <sup>-20</sup>
SrCO <sub>3</sub>	5.60 × 10 <sup>-10</sup>	Sr(OH) <sub>2</sub>	3.20 × 10 <sup>-4</sup>
ZnCO <sub>3</sub>	1.46 × 10 <sup>-10</sup>	Zn(OH) <sub>2</sub>	3.00 × 10 <sup>-17</sup>

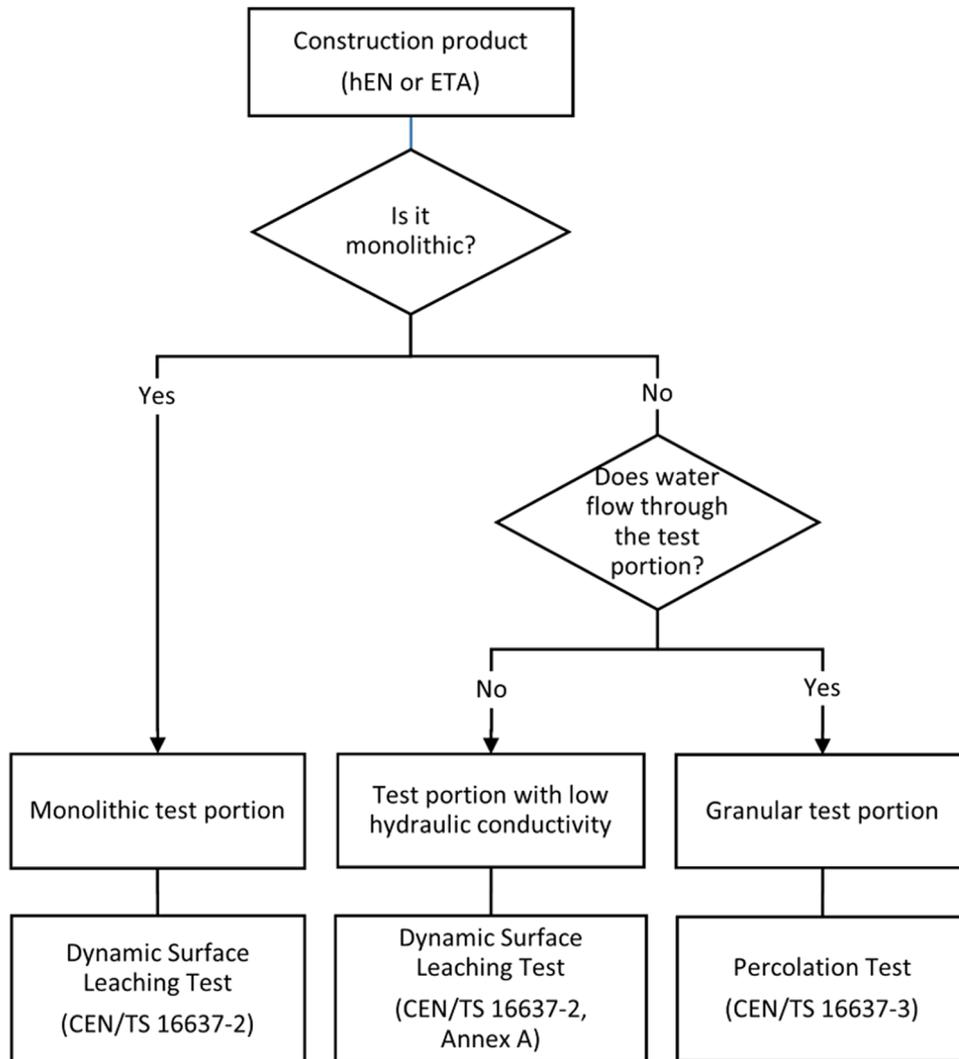


Fig. A1. Determination of leaching test methods for construction products (DIN EN 16637-1, 2024).

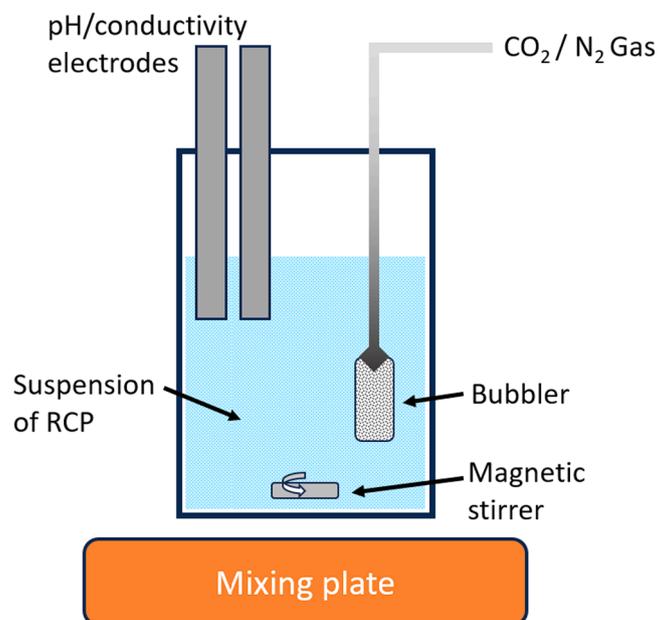


Fig. A2. Sketch of the enforced carbonation experimental setup (Zajac et al., 2020a, 2020c).



Fig. A3. Pictures of enforced carbonation reactor installed in a cement plant in Poland (Beumelburg, 2025; Zajac, 2025).

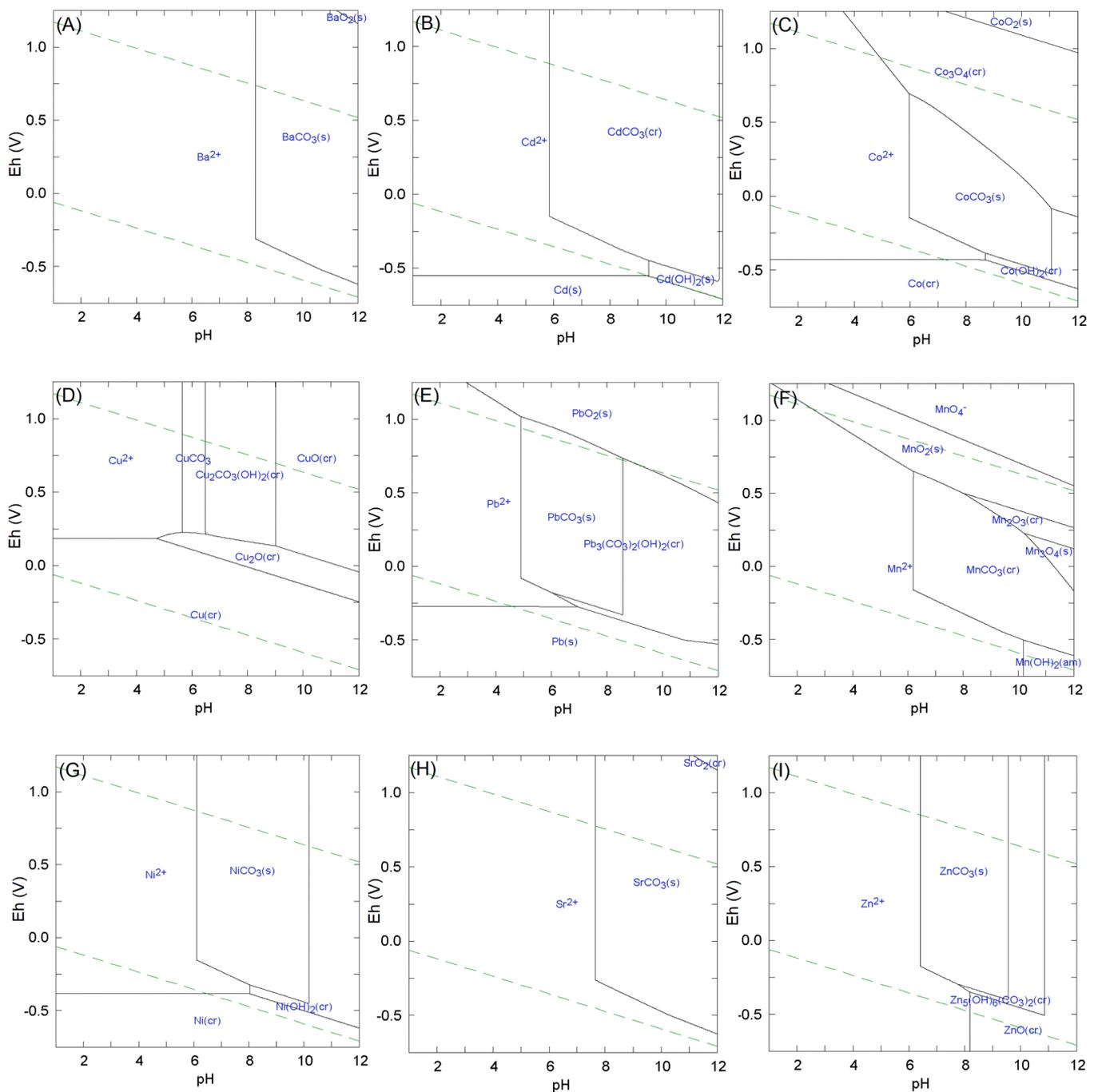


Fig. A4. Pourbaix diagram of divalent metals  $M^{2+}$  in the  $CO_2$ - $H_2O$  system at 25 °C, generated using Medusa-Hydra software (Puigdomenech, 2004);  $[M^{2+}] = 1.0 \times 10^{-5}$  M,  $[C]_{Total} = 5.0 \times 10^{-2}$  M; The  $[C]_{Total}$  was based on the total inorganic carbon in carbonation solution as calculated by Zajac et al. (2020b).

## Data availability

Data will be made available on request.

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