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Opening Letter of RILEM TC UMW: Upcycling Powder Mineral Wastes into Cement Matrices — Challenges and Opportunities

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Abstract

The cement and concrete industries are currently facing the urgent and arduous challenge of decarbonisation and material circularisation for improved resource efficiency. The pursuit of new raw materials and binders that will improve sustainability is urgent, especially as end-of-pipe carbon capture and storage (CCS) technologies have not yet been scaled up economically even after five decades of research and large investments. On the other hand, society is facing the colossal issue of managing mineral wastes which are produced in several Gts per year globally, posing a massive environmental and societal liability. Many of these mineral wastes have elemental and mineralogical profiles that make them good candidates for use as clinker raw feed or supplementary cementitious materials. Although the published research on the topic is extensive, it is not organised, lacking a systematic comprehensive approach, making valorisation challenging. RILEM TC UMW was developed to address this gap and create a framework for realising the potential of upcycling mineral wastes focusing on using powders as either clinker raw feed or other binder applications while excluding discussion on calcined clays and mineral carbonation.

Keywords: Upcycling; Mineral waste; Low-carbon clinkers; Supplementary cementitious materials; Alkali-activated materials.

1 Introduction

The utilisation of minerals currently forms the backbone of most technological developments in all sectors of our socioeconomic environment. The shift towards more sustainable energy production technologies including, e.g., wind turbines, solar panels, and energy storage (batteries) will lead to intensified mining activities in the years to come as we extract the minerals needed to produce the related devices [1]. Moreover, recent estimates predict that the construction sector will significantly contribute to the growth in raw materials consumption for the next few decades [2]. The increase in mining and consumption of raw materials through urbanisation and growth of consumerism generates vast volumes of mineral waste, and the management and utilization of such wastes are becoming an increasingly urgent issue in the global sustainability landscape with significant environmental and social challenges including biodiversity, climate change, and quality of life for humans [3, 4]. For example, construction and demolition waste (CDW) accounts for more than 30% of all waste generated in the European Union [5], and could be instrumental in achieving material circularity in concrete production if/when waste concrete crushing technology and separation of the paste from the aggregate fractions is matured and scaled up. The problem of mineral waste generation is global and is not restricted to certain geographical regions or country development levels. What changes case by case are the types and volumes of waste generated.

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Mineral wastes are typically rich in SiO_2, Al_2O_3, CaO, MgO, and Fe_2O_3, which makes them very attractive for utilization and upcycling in cement-based materials. This can be either as alternative materials used in clinkering, as alternative supplementary cementitious materials (SCMs), or used as precursors in chemically activated cement. It is worth highlighting that traditional SCMs face several challenges concerning their continuous availability, due, e.g., to the "decommissioning" of the industrial processes they originate from as a waste/secondary raw materials. [6] This highly prioritizes the need to shift to new types of cementitious materials, thus justifying the need of exploring and expediting the use of alternative sources. The sustainable transition in cement/concrete industries roadmap published by the International Energy Agency has, as a top priority, the transition to new cementitious binders by 2035 [7], a priority that echoes the report on cement decarbonisation by the European Cement Association [8], and the Institution of Civil Engineers low carbon concrete route map [9, 10].

For the construction sector and more specifically for cement–based materials, there is a great opportunity ahead. Being a high-volume industry, cement/concrete manufacturing contributes heavily to carbon emissions and is one of the largest consumers of natural resources. Adopting extensive circularity frameworks that will enable the upcycling of potentially useful mineral waste can lead to significant improvements. Recent reports estimate the market growth potential between 80 and 110 billion euros [11, 12].

This paper discusses the current landscape in the context of the scope and focus areas of the RILEM Technical Committee (TC) on Upcycling Powder Mineral "Wastes" into Cement Matrices [13]. Within its 5-year lifetime, this TC, aims to develop a robust roadmap for the swift and widespread adoption of the upcycling of mineral wastes in cement and concrete in the short-to-midterm. The focus is exclusively on fine fractions (powder or powdered like cement) and their contribution to the development of the microstructure and performance of cementitious matrices. Calcined clays and mineral carbonation are excluded from this contribution as they are out of the scope of this TC.

2 Current landscape

Research for identifying new alternative mineral additions has gained considerable interest during the last decades. We focus on materials other than traditional SCMs and mineral additions whose use is consolidated in cement standards (e.g., ASTM C150/C150M, EN 197-1), such as coal fly ashes and ground granulated blast furnace slags. Among recent developments in the area, extensive studies are reported on the use of industrial and agricultural ashes [14] as SCMs, while some progress is also reported on metallurgical residues, other than GGBFS [15]. The use of fines from recycled concrete is also under investigation for use as SCM [16] or constituent of clinker raw meal [17] and has been incorporated already in the European cement standards (EN 197-6). Mining waste is an extremely variable material in mineral composition, needing specific approaches depending on the target resource and local geology. For example, due to the association of coal seams with kaolinite, mining residues in the coal industry are often appreciated for use in cementitious materials, such as Coal Mine Waste Geomaterials (CMWGs), especially after thermal activation into a metakaolin-rich material [18-21]. Subsequent section 4.1 discusses clinkering processes in more detail.

Despite some recent advancements, the issue of managing considerable volumes of mineral wastes remains unresolved. As listed by Snellings et al. [15] and calculated by Peys et al. [22] for metallurgical residues and Pires Martins et al. [23] for mining residues , the approximate global yearly production of some potential new alternative minerals is: 200 Mt steel slags; 60 Mt from non-ferrous metallurgy slags, 170 Mt processed bauxite, and 5 to 10 Gt mine tailings. While the 5-10 Gt comprises only flotation tailings, the whole range of mining and quarrying activities is estimated even at 20 Gt/year [24]. In some of these cases, a small percentage of the mineral residues is used as filler in cement or concrete productions. The remaining of the wastes are either downcycled or not being used and therefore must be managed accordingly. These are stockpiled, imposing land use and environmental liabilities. Estimated current volumes of stockpiled materials are for instance 10.7 Gt in the coal industry [19] or 55.7 km^3 in tailings storage facilities [25]. Mining and metallurgical residues contain in some cases still sizable amounts of critical raw materials (CRMs) that were lost during initial processing. Also, the metallurgical sectors yearly generate a substantial volume of mineral wastes. Some of these residues are expected to increase further in the coming years, such as byproducts from the production of lithium hydroxide, which is key in battery manufacturing. These pyrometallurgical processing residues are primarily composed of aluminosilicates and calcium oxide which can be used in the cement industry as raw material for clinker production or as an additive for cement, replacing conventional precursors and SCMs. Pretreatment or beneficiation processes should (and in some cases must) be considered to recover these valuable elements and/or provide a more suitable version for use in cementitious materials.

These considerations even still exclude residues such as dredged sediments and excavated soil, for which, due to the overall lower metal content, beneficiation processes consider rather thermal treatment for increasing the reactivity and decreasing organic matter content [26, 27]. In terms of scale, dredged sediments and excavated soils are unavoidable in the discussion, with recent figures showing more than 600 million m^3 of sediment being dredged globally per year [27]. Excavated materials are even estimated to have a 5 times larger yearly volume [28].

Currently there is comparably very little application of mineral waste in cements. This can be attributed to several aspects including: 1) the mineralogy and/or minor element composition of some wastes can lead to unfavourable effects in a cementitious matrix such as delayed or flash setting, reduced strength and durability issues [29-31]; 2) there is globally no systematic research on this area as the abundance and composition of mineral wastes are very location-dependent, for several reasons including natural differences in geology and local legislation and know-how. While trying to globally enforce focus on more mainstream sources, the local character of many raw materials should not be overlooked; 3) limitations still hold on advanced and swift characterisation and testing techniques; 4) legal limitations; 5) limitations in relation to standardisation, building codes and social acceptance aspects regarding the use of novel constituents and binder types; 6) contaminant leaching and/or human exposure (e.g., asbestos; radioactivity).

Consequently, these mineral wastes are not well understood and not properly mapped against their potential applications. While some advancements have been achieved in developing new cementitious materials with their market maturity being quite high [15], the large volume of marginally used mineral waste should not be ignored. Currently there are barriers associated with their upcycling, and hence a systematic widespread research activity is required. The advancements of the last few decades in post-processing techniques and performance testing provides us with tools to do so; and we believe that knowledge technology transfer from other sectors is lacking and must be supported and encouraged. That is the scope of the RILEM TC UMW.

3 Challenges

3.1 Identification and mapping

To appropriately and responsibly use mineral waste as cementitious materials, knowledge on the quantitative and geographical availability, and general chemical and physical properties is required. Unambiguous nomenclature for a certain material is needed, and the variability in composition that can occur between similarly named materials necessitates an organized categorization of mineral wastes as well as an understanding of procedures to systematically blend and create an economically adequate/viable feedstock. To this end, identification of the residues that are available for use in cement and arranging them into a mineral waste "family tree" is the starting point of TC UMW. The challenge lies in being sufficiently broad to be relevant for as many cases as possible and, at the same time, being sufficiently detailed to provide exploitable information. Existing databases that could be used as an example or starting point are for instance the resource databases listed by Mwiti Marangu et al. [32].

Once it is clear which materials might be considered relevant for use in cement, their technical properties and performance assessments in cementitious systems can be carried out in a structured way. Although industries do not typically report annual data on waste production and its quality for cementitious systems, the volume of industrial by-products can be estimated. This estimation is based on the yearly production of the finished products that generate the waste as well as legacy/landfilled materials. Such data can be retrieved from several sources, including, e.g., industry reports, scientific literature, textbooks. The mapping of composition and properties in cementitious systems needs to incorporate enough references/surveys to enable an assessment of company/industry/location specific quality, versus general conclusions about a certain type of mineral waste.

3.2 Characterisation

Swift and accurate chemical and mineralogical analysis is crucial to evaluate the potential for a mineral waste to be used in cementitious materials as well as to understand feed corrections needed. It is timely that technological advances in materials science have allowed the necessary, reliable, fast and portable characterization instruments to come to market. Regarding the use in clinker raw meals, an accurate assessment of the chemical oxide composition is generally sufficient, but in some cases, the mineralogy, oxidation state, polymorphism of the mineral wastes, as well as their particle size will affect their use in cements and subsequent performance [33]. On the other hand, the valorisation of specific wastes, without substantial modification of their phase assemblage, in SCMs and chemically (alkali or acid) activated materials, requires that the phase composition is accurately quantified [34]. An accurate use of mineralogy determining techniques such as X-ray diffraction (XRD) to investigate in more detail the reactive phases are necessary to increase understanding of the waste characteristics, reactivity with portlandite, acids, or alkaline activators as well as indicate towards leaching, and durability performance, and to enable the proposition of robust processing/valorisation schemes from waste to cement. This requires a careful estimation of: a) the reactivity at the solid-fluid interface and in aqueous solution of individual crystalline phases present in the specific residue; and b) the amount and chemical composition of the amorphous phase. Guidelines for this can be found in the general cement literature, such as the book by Scrivener, Snellings, and Lothenbach [35]. More specialized literature can be consulted for more specific mineral wastes. The characterization of Mg-rich [36] or Fe-rich materials [37] can provide additional challenges and many mineral wastes fall into these categories. For example, the high paramagnetic nature of Fe hinders the use of nuclear magnetic resonance (NMR) spectroscopy and, in such cases, other techniques such as Mössbauer spectroscopy might become more attractive.

Mineral wastes may also have large inhomogeneity/variability and hence are prone to sampling errors during their characterization. Depending on the process of origin and details of the storage facilities, mineral wastes sampled at different spots in a waste heap can have variable chemical composition, phase composition, contaminant concentration, and particle size distribution. Thus, development and utilisation of rapid characterisation tools such as hand-held X-ray fluorescence (XRF), Raman spectroscopy, Laser-induced breakdown spectroscopy (LIBS), and Dynamic image analyses (DIA) for particle size and shape characterization are necessary for determining the "waste" material profile, which would enable the engineers to choose a beneficiation and upcycling pathway. Recently, Samouh et al. [38] have shown that a hand-held Raman with the assistance of photo-bleaching could be more effective than lab-based XRD in identifying certain minor mineral phases in municipal solid waste incinerator (MSWI) ashes. Even when the phases in the waste material are homogeneously distributed, the mineralogical characterization by XRD is complicated by the presence of crystalline and amorphous phases with possible peak overlapping (due to the substantial amount of minor phases in e.g. bauxite residue [39] and mine tailings [23]), solid solutions (e.g. Al and Cr incorporation in goethite and hematite in bauxite residue [39] or Mg and Ca incorporation in fayalite in non-ferrous slags [40]), and preferred orientation-sensitive phases. These issues can be partially mitigated by an integrated characterization approach that combines complementary characterisation techniques, such as XRF, Energy Dispersive X-ray (EDX) analysis, Raman spectroscopy, and XRD. This TC will also recommend specific characterisation techniques based on the mineral waste and upcycling pathway.

3.3 Regulatory framework

The pursuit of sustainable construction materials requires navigating a complex regulatory landscape involving both technical and legal aspects. Although this topic is crucial, there is a noticeable lack of comprehensive research that pulls together the various regulatory frameworks and their impact on using mineral waste in low-carbon cements, especially across different regions. Even though the discussion here is focused on EU construction regulations, more work is needed to expand the evaluation regulatory hurdles in other key regions. Nevertheless, the EU regulatory framework is discussed here as a representative example, with the understanding that the principles and challenges addressed are also of relevance to the broader international scientific and technical community.

According to Directive (2008/98/EC) of the European Parliament and of the Council on waste, 'waste' means any substance or object which the holder discards, intends to, or is required to discard [41]. For mineral waste-derived SCMs to be reclassified as by-products, they must meet criteria like further use certainty and compliance with environmental standards. However, navigating these classifications is just one part of the regulatory framework and additional product-specific regulations further ensure that sustainable construction materials meet rigorous safety, environmental, and performance standards. Chemical, product, and structural regulations apply to primary materials used in construction. REACH (Registration, Evaluation, Authorisation, and Restriction of Chemicals) and CLP (Classification, Labelling, and Packaging) regulations ensure environmental and human health protection by assessing chemical risks and communicating hazards [42, 43]. The Construction Products Regulation (CPR) requires SCMs to meet essential criteria like mechanical resistance, stability, fire safety, and environmental protection, with compliance demonstrated through a 'declaration of performance' and CE marking [44]. Testing protocols and standards (e.g., EN, ASTM) ensure construction materials meet safety, environmental, and performance standards. However, their prescriptive nature can hinder novel materials. Recent updates show standardization bodies are becoming more lenient towards new materials, recognizing the need for new resources, provided performance and durability are well documented [45].

Mineral wastes may also contain, to varying extents, some Naturally Occurring Radionuclides (NOR) [46], and their upcycling into construction and building materials raises radiological protection issues specific to their nature [47, 48]. This has also been reflected in current EU directive [49], which takes into account the specificity of exposure to NOR with respect to artificial radioactivity. In any case, the development of a tailored assessment framework to enable operators to recover and reuse NOR, also in the construction industry, is recognized as crucial to avoid NOR accumulation which would result into an even worse exposure to humans and the environment [50].

Transitioning to recycling in the cement and construction industry raises key questions. Despite REACH exemptions for cement clinker, new waste-derived SCMs face stringent safety assessments [51]. A major issue is defining safe use for waste products, with no EU-wide end-of-waste criteria for mineral waste-derived SCMs, aggregates, or fillers. This lack of harmonization causes confusion, as national frameworks have varying chemical constituent limits, leading to discrepancies across member states. Various national frameworks enforce different limit values for chemical constituents that may pose environmental or health risks, leading to significant discrepancies across member states. Snellings et al. [15] reports on acceptable ranges of heavy metals for cement and concrete applications in Germany, Switzerland Netherlands, France, Flanders and Austria. This is while countries like Sweden have only general recycling limits for application in unbound construction which is tremendously lower than the values reported by Snellings et al. [15]. Armistead and Babaahmadi [52] have summarized the limits (lower and higher limits) associated with several national end-of-waste recycling criteria, showcasing the high scatter between the data. In this context, leaching tests, both percolation-based (CEN/TS 14405 and CEN/TC 351/TS-3) and batch-based (EN 12457-1, 2 or 3), are critical in evaluating the potential risks of using mineral-based waste-derived SCMs.

Depending on the country, not only the limit values are different, but also the proposed test methods vary; while some countries pose hard limits on the total content of contaminants, others pose limits on the leachability via batch leaching or column leaching tests on the mineral waste or a concrete specimen made with the mineral waste [53]. In the work by Snellings et al. [15] the limits in some countries are reported only based on total solids (Germany and Switzerland), while in some other countries Netherlands, France, Flanders and Austria leaching limits exists. The relevant leaching tests for these limits are however, not referred to by Snellings et al. It should be noted that these leaching tests primarily focus on the granular, ‘unbound’ states, while “bound” or application of so-called monolithic samples would be more appropriate. Currently, only the non-harmonized CEN/TS 16637-2 specification exists for monolithic leaching tests. Advancing mineral waste-based materials in construction requires systematic documentation of heavy metal contents in SCMs, suitable leaching tests, and investigation of heavy metal dissolution. Discussions should also cover additional mineral waste-related legislation.

Achieving a balance between efficient use of mineral waste in low-carbon cements and ensuring long-term environmental quality is essential for sustainable construction.

In addition to concerns about unwanted elements and radionuclides inherent to the material, respirable crystalline silica (RCS) poses a significant occupational health risk during handling and processing of silica-containing mineral waste. While the bulk material may not fall under specific restrictions in EU chemical regulations such as REACH (Regulation EC No. 1907/2006) [54], which governs the safe marketing of substances, the generation of respirable dust during use, such as grinding or mixing, activates a separate regulatory framework focused on workplace exposure. In the EU, RCS released through work processes is classified as a carcinogen under Directive (EU) 2017/2398, amending Directive 2004/37/EC, and subject to an occupational exposure limit of 0.1 mg/m^3 (8-hour TWA) [55]. As such, employers handling these materials must implement appropriate dust control, monitoring, and health surveillance measures to ensure safe working conditions. Comparable requirements exist internationally, for instance under OSHA regulation 29 CFR 1926.1153 in the US [56].

3.4 Beneficiation techniques

Some mineral waste materials are not suitable by default and require processing to enable an optimal use in cementitious materials. To enable valorisation of mineral wastes in specific binder systems, substantial research effort is directed to pretreatment and beneficiation techniques. The problems to be solved with such treatment can vary between residues, but environmental compatibility and reactivity of the material are the two most investigated [15, 22]. The process portfolio to increase the quality on these aspects seen in literature comprises the up concentration of certain compounds by physical separation, chemical processing, and thermal treatment [57-59]. There has been significant progress in physical separation processes, such as comminution and associated technologies, which are fundamental, for example, in the management of CDW [60]. These include, e.g., impact crushing, magnetic separation, electric pulses, or microwave heating [61-63]. Chemical processing via dissolution or digestion has been regularly used, for example, through washing of salts found in mineral wastes such as cement kiln bypass dust [64, 65] and aluminium salt slag [66] as well as in the leaching of heavy metals [67]. Thermal treatment is also well-known for increasing the reactivity of, for example, clay-containing wastes, such as dredged sediments [26] or coal mining waste [68], for use as SCMs or cement precursors.

A holistic flowsheet composed of a toolbox of physical separation (mineral processing) and metal extraction and recovery processes, as shown in Figure 1, may be necessary for mineral wastes which have high contents of potentially hazardous metals or metalloids [69]. In general, the beneficiation techniques currently utilised in the industry mostly focus on removing heavy metals or other undesired chemical elements from the materials before utilisation (or to ease storage). However, in some cases, it may be possible to modify the phases, at source, containing unwanted elements thus capturing or immobilising them. For example, chlorellestadite formation in MSWI fly ashes can immobilise chlorine potentially enabling usage in steel-reinforced concrete [70], while reducing the leaching of heavy metals such as Pb through crystallo-chemical incorporation is also possible [71].

On the other hand, beneficiation techniques might be used for materials which were previously considered to be off-spec. With the steady closure of coal powerplants and introduction of low-NOx burners in the western countries, there has been a shortage of high-quality coal fly ashes in Europe that meet industry specification for high-value SCM usage [72]. This has created a relatively large market on beneficiating ashes from the low-NOx burners or from ash ponds (legacy ashes). These ashes generally have a higher carbon content, and different techniques such as electrostatic removal, wet separation, and size-fractionation (large fly ash particles are generally richer in carbon) are being employed for removing the high carbon ash particles [72]. More research and development activities are expected in the industrial scale to further lower the energy and cost of these techniques, while knowledge exchange from other industrial sectors into the cement industry can and should be enhanced.

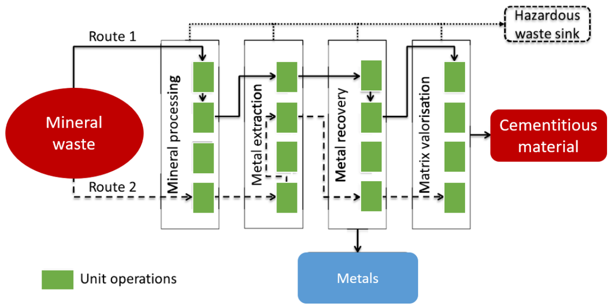

Figure 1. Toolbox of processes to design a flowsheet for the beneficiation of a mineral waste, modified from Spooren et al. [67].

4 Opportunities

4.1 Clinkering

Upcycling waste minerals through clinkering depends on the chemical compositions of the mineral wastes but has the benefit of not relying entirely on mineralogy (and e.g., oxidation state). The high temperature processing conditions facilitate a drastic transformation of the mineral composition, enabling the utilization of some residues that are too unreactive for use as SCM or in chemical activation. The chemical composition limits the maximum amount of waste material that can be incorporated/replaced in the raw meal of manufacturing cement clinker [73]. The options widen when considering alternative clinkers to PC as using these materials for manufacturing alternative clinkers often allows for using them at a higher percentage of raw meal feed compared to PC. For example, sulfur-rich mineral waste can be efficiently valorised in calcium sulfoaluminate or sulfosilicate based cement [74, 75] (where the processing conditions do not favour the release of SO_2 gas into the atmosphere), mineral wastes containing chlorine can potentially be upcycled through alinite based cements for unreinforced concrete applications [76] (where the chlorine is locked in target mineral phases), and high iron-containing waste can be upcycled through manufacturing high ferrite or oil-well cements which have special properties [77, 78].

Many waste minerals such as steel slag and some mine tailings [79] contain high quantities of Ca, that is not in carbonate form, but in silicate or oxide form. Using these mineral wastes as raw materials for clinkering reduces the need for limestone calcination and thus reduces the carbon footprint of cement manufacturing [80]. This presents a unique opportunity to combine multiple improvements in today's cement manufacturing - resource-efficiency, lower CO_2-footprint, landfill diversion; while reducing the cost of manufacturing and promoting circularity. Similarly, Si-rich mineral wastes containing a significant amount of Ca, such as recycled concrete fines, can be utilised to diminish the CO_2 footprint of the clinkering process [17]. This has recently also been demonstrated in co-production of steel in an electric arc furnace [81].

Mineral wastes contain different minor elements (Mg, Mn, Ti, Cr, Zn etc.), and these can affect the performance of the clinker, both in terms of processing conditions (e.g. burnability, emissions) and performance of the end-products (cement and concrete) both during service life and end-of-life. In certain cases, these minor elements may improve the performance and processing conditions. For example, incorporating Mn in high ferrite cement has been shown to improve hydraulic reactivity [82], whereas F in the raw meal can reduce the clinkering temperature [83]. Identifying the minor element(s) that can improve the clinkering processing conditions or end-product performance presents a unique opportunity to encourage upcycling waste minerals for clinkering. High temperature thermodynamic modelling [84-86] is an essential part of this discussion in the efforts of assessing suitability of certain minerals to be part of clinkering processes.

Safety and sustainability, by design, of clinker/cement formulation to form certain phases (such as ettringite or Friedel's salt) upon hydration, should also be considered while upcycling waste materials containing unwanted chemical elements into clinkers, as some hydration phases are more stable and can immobilise many unconventional or unwanted chemical elements from leaching [87-90]. Furthermore, the targeted formation of non-hydraulic phases such as spinel can prevent the leaching of deleterious chemical elements [91, 92].

4.2 Use as supplementary cementitious material

The technical performance of mineral wastes as SCM is determined by the mineralogy. This determines the solubility in the conditions posed by the cement pore solution and the participation in hydration reactions. Many SCMs contain a reactive aluminosilicate phase, which is typically amorphous [15], but might also contain substantial amounts of calcium and iron or even a minor amount of magnesium [93]. This reactive fraction may consist, for instance, of a meta-clay phase originating from the calcination of a clay-containing waste [26], a glassy phase in a slag originating from a pyrometallurgical process [94], or a silica gel forming as a by-product of the carbonation of a calcium silicate (hydrate) phase [16, 95]. In exceptional cases, the reactive silicate is not amorphous; crystalline calcium silicate phases such as belite can also contribute to the hydration reactions, as well-studied in cement chemistry. Examples of mineral wastes that are appearing as emerging SCMs or potential SCMs of the future are highlighted in Figure 2, comparing their reactivity as SCM with several competing SCMs.

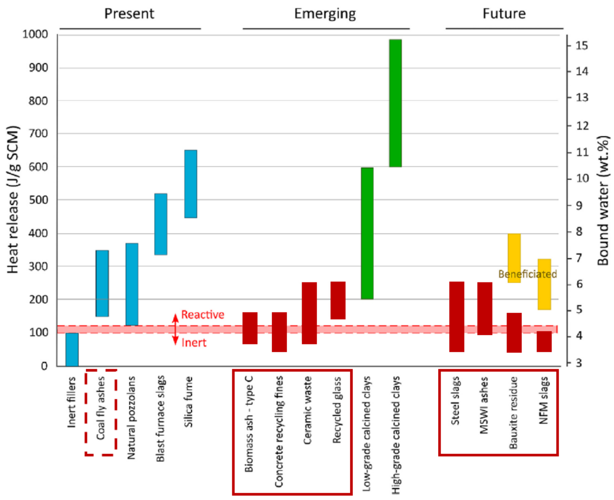

Figure 2. R^3 heat release as a measure for reactivity of emerging and future SCMs, modified from Snellings et al. [15], highlighting mineral wastes in red. Coal fly ashes are highlighted with a dashed line, as whether they are discarded or subject to shortage depends on the region.

Minor components in mineral wastes might have a significant influence on the hydration of cement phases. Organic matter and organic acids can cause substantial delay of the hydration reactions [27, 96], but also the presence of heavy metals can decelerate cement hydration [97], while alkaline mineral waste might cause extensive acceleration [98].

Environmental quality is an important aspect when dealing with residues. The content and leachability of potentially harmful substances in concrete needs to be safeguarded and tracked. Depending on the origin of the waste, contaminants of organic origin, metals and metalloids, or potentially asbestiform amphiboles or serpentines can be present. Substantial research efforts are directed to study whether these wastes need to be avoided or whether beneficiation processes exist, or can be developed, that can convert, destroy, or immobilize the contaminant.

4.3 Chemical activation and use as activator source

Chemical (alkali and/or acid) activation is a versatile process by which wastes of different nature can be made reactive, inducing the precipitation of reaction products that convey cohesive properties to the construction material incorporating them. Ground granulated blast furnace slag has notably been used as a reactive powder to produce alkali-activated binders, with application in structural concrete starting from the 1960s [46]. More recent applications of alkali-activated materials (AAM), comprising waste-based AAM can be found in a recent report [47]. Given the enormous chemical and mineralogical variability of waste materials that can potentially be used for chemical activation; thorough characterization is essential for defining an appropriate activation strategy.

Waste materials for alkali activation can be classified based on whether they contain reactive Ca or not, since the concentration of Ca^{2+} in aqueous solution can, but not always, determine the specific reaction pathway. This can lead to the formation of reaction products with calcium (C-A-S-H) providing charge balance and network modification or stoichiometric Na as charge balancing cation (N-A-S-H), the latter being able to accommodate limited amounts of Ca in its structure. An optimal formulation of AAMs requires that the bulk composition (reactive fraction of the utilised waste and activator) is compatible with the target stoichiometry of the reaction products (see Figure 3), e.g. to avoid that excess alkalis react with atmospheric CO_2 to form carbonated phases that may potentially damage the microstructure. As an example, within the sub-class of low-Ca AAM, rice husk ash represents a Si-rich highly amorphous (and reactive) waste. However, its high solubility in sodium hydroxide and sodium silicate solution does not guarantee an appropriate setting and hardening because the system will be Al-deficient and hence undersaturated in N-A-S-H. Subsequently, sodium aluminate represents a more suitable activator for this specific waste (and in general for wastes having a Si-dominated chemical composition of the reactive fraction) [49].

Alkaline activation can also be an efficient strategy for the utilization of Fe-rich waste, generally characterized by limited reactivity in aqueous solution. Literature studies of Fe-rich AAM materials have shown a large variability in mechanical performance [99] and, again, an accurate chemical and mineralogical analysis is fundamental to define an appropriate activation strategy and achieve an acceptable performance. This also requires a better understanding of the role of Fe in the dissolution-precipitation reactions that can follow complex reaction pathways, which are different from N-A-S-H and C-A-S-H formation [100]. Rice husk ash [51], maize ashes [101], and other Si-rich residues, such as waste glass and silica fume [52], were identified as suitable secondary materials to produce sodium silicate solutions, leading to a significantly reduced environmental footprint without compromising performance. Identifying other locally available wastes for alkaline activator production should be encouraged, considering that most of the environmental and economic cost of alkali-activated materials is associated with the alkaline activators. Therefore, the deployment of this technology may strongly benefit from a circular approach.

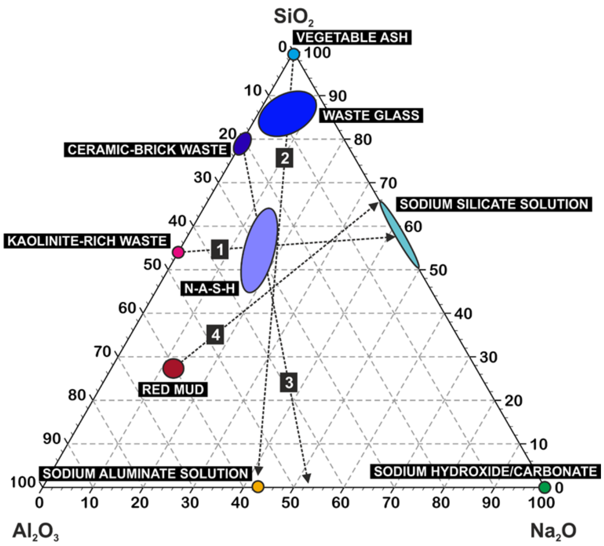

Figure 3. Possible activation strategy of selected mineral wastes: 1) Al-rich wastes can be appropriately activated by sodium aluminate solutions with SiO_2/Na_2O ratios in the range 1.2-1.8. Appropriate dosage of the activator is needed to avoid excess Na in solution, leading to the formation of alkali carbonates resulting in efflorescence. 2) Al-poor wastes such as vegetable ashes, waste glass and ceramic waste request an Al-bearing activator for the pore solution to become saturated in N-A-S-H. The addition of NaOH (3) can be used to regulate the concentration of Na and pH. (4) Wastes having a relatively low concentration of Si may request sodium silicate solutions having a slightly larger Si concentration, although a too high SiO_2/Na_2O would decrease the pH and activating power of the solution. N-A-S-H stability field taken from [16, 102-104].

5 Closing remarks and ways forward

The construction sector, traditional mining and metallurgical industries, and the wide adoption of sustainable technologies are associated with the yearly generation of mineral wastes at Gt-scale. Despite changing energy and manufacturing technologies while chasing a CO_2-neutral society, there are no prospects of significant decrease of the overall volume of mineral waste in the short to mid-term. Such a development would also require the organization and deconvolution of supply chains, for instance based on criteria of decentralisation and industrial symbiosis where possible. This has a considerable impact on two significant global problems: the decarbonisation of cement production and the management of waste.

The cement and concrete sector sources materials from a relatively small area close to source and to reduce transportation costs, and the mineralogical composition of wastes often have a high geographical and temporal variance. Thus, there would be no single solution optimal for all regions on the planet. Local materials must be thoroughly characterised and depending on their mineralogical, elemental, and chemical characteristics they need to be assigned for optimal use in binder systems; while a systematic overall methodology and frameworks would support this endeavour. Further, striving for zero CO_2 emissions by 2050 will impose the need for novel processes and approaches but limiting our vision to 2050 may also cause problems for future generations to solve. Alternative limestone decarbonation processes [102], carbonation curing [103], the production of SCMs by carbonation [16], or magnesium-based cements [104] are some examples of newly developed solutions which can be utilised towards developing a framework for large scale valorisation of upcycled mineral wastes; yet, the world population and demand for resources may eventually balance, enabling the prospect of a truly circular approach and the diminished use of virgin raw materials

In summary, the purpose of this contribution was threefold. First, to discuss the current situation in relation to the growing production of mineral wastes globally. Secondly, to highlight and consolidate the research done and the research needs in relation to upcycling mineral wastes and, finally, to emphasise the need for steep and radical changes. Various stakeholders in this discussion have created mission statements and RILEM TC UMW aims to integrate the efforts of several experts from different parts of the world towards achieving positive change. The TC has detailed 5-year working programme which will be delivered by four working groups that will: (i) develop mineral wastes inventories/maps and classification; (ii) explore utilisation of mineral wastes in clinker-derived cementitious matrices; (iii) investigate utilisation of mineral wastes in alkali-/acid- activated cements and (iv) discuss environmental considerations and wider regulatory frameworks. The anticipated body of work that will be developed from this TC is expected to inform future research directions in the field.

While the TC UMW aims to consolidate existing information for swift adoption of technologies; there are considerable and significant considerations and challenges (beyond TC UMW) associated with the use of alternative raw materials or alternative binders, which mostly centre around "safety and sustainability by design":

- Recycling potential: If a mineral waste must be reused into construction, an appropriate plan for its end-of-life must be worked out. Reuse/recycle approaches must be considered at early stages, not to incur the risk of postponing a problem and handing it over to future generations. Furthermore, genuine alternative binders and even blended cements are currently being developed with no proposition to fully recycle. Recycling will also need to consider accumulation of deleterious elements in our building stock.
- Redesign: Beneficiation can, in many cases, be carried out at source through understanding and planning of the second, or infinite lives of the materials in question. Approaches such as design to recycle, and industrial symbiosis have already emerged but require considerable effort across industry and disciplines to not only merely engage downstream but to get involved in grassroot operations. Knowledge sharing and transfer across industry and from academic institutions will be key to widening these benefits and continue working together towards a better society.
- Health and safety: It is imperative to prevent the dispersion of environmentally hazardous materials into the built environment. That is very relevant for engineered compounds such as mineral wastes that contain asbestos or heavy metals. Some environmental and health effects of the mineral wastes considered in this paper have been shown in some instances, due to the release of heavy and toxic heavy metals and even radionuclides, resulting in cost burdens that can even become socialized in underdeveloped or developing countries. An environmental and health impact analysis combined with remediation cost-benefit analysis should be conducted to understand the environmental and socioeconomic costs and benefits associated with any waste management or tailing body and associated reuse. This also calls for appropriate legislation for safe use of the secondary raw materials which can be obtained from the valorisation of these mineral wastes.

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Declaration of interest

T. Hanein is shareholder of United earth4Earth Holding. A. Baral is one of the inventors on the U.S. patent application 63/614,816.

Authorship statement (CRediT)

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