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Coal mining wastes valorisation as raw geomaterials in construction: A review with new perspectives

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25 October 2021

To: Professor Jiří Jaromír Klemeš,
Editor-in-Chief, Journal of Cleaner Production

RE: **Coal mining wastes valorisation as raw geomaterials in construction: A review with new perspectives**

by Thanh Liem Vo, William Nash, Marco del Galdo, Mohammad Rezania,
Rich Crane, Mohaddeseh Mousavi Nezhad, and Liberato Ferrara

Dear Prof. Klemeš,

My co-authors and I are happy to submit the above paper to the Journal of Cleaner Production for possible publication.

In this contribution experts from different disciplines joined forces to provide a state-of-the-art review for practical uptake of coal mining waste geomaterials in construction engineering, particularly for higher level construction applications. The areas of expertise of the authors include a wide range, from geochemistry, material sciences, hydro-geology, and geomechanics, to constructions, concrete technology and structural engineering. This enabled us to present a comprehensive review and provide a multi-angle understanding of different properties of coal mining waste geomaterials to assess their suitability as replacement for both aggregates and binders in concrete and cementitious composites. The review also covers the interactions between mineralogy, geotechnical indices, and state-dependant properties of mining wastes together with the implications of their application.

This paper will become paper of reference for our current and future applied research on valorisation of mining wastes, and most possibly that of other researchers working in resources recycling and/or sustainable construction fields. We hope that you and the reviewers will find this paper worth publishing in your journal. If you should need additional information, please contact me.

Yours sincerely,

M. Rezania

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cc: Dr. Vo and Prof. Nezhad at the University of Warwick,
Dr. Nash and Dr. Crane at the University of Exeter,
Dr. del Gado and Prof. Ferrara at Politecnico di Milano.

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Coal mining wastes valorisation as raw geomaterials in construction: A review with new perspectives

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Abstract

Historically coal mining wastes have been viewed as heterogenous and hazard-prone geomaterials. Given that failures of colliery tips and tailing dams are reported on a regular basis, reclamation of coal mining wastes from storage facilities is increasingly being considered. There is a resistance to the use of coal mining waste in construction industry despite scattered but growing reports of successful applications. As the construction industry around the globe seeks to reduce its carbon emissions by looking for supplements for cement, the voluminous amount of coal mining wastes currently stored in spoil heaps and impoundment facilities present a potential source of raw materials. This article reviews the literature on the geochemical, geotechnical and structural engineering properties of coal mining waste geomaterials to assess their suitability as replacement for both aggregates and binders in concrete and cementitious composites. It is found that coal mining wastes are indeed good candidates (as raw materials) for the uptake and process into higher level construction purposes. Geochemically, the key to a successful upcycling operation is the knowledge of their mineral contents (which is typically diverse and varies from one mine to another) and the processes they undergo while being transformed into constituents of new materials. From a concrete technology perspective, the key to a successful utilisation is the mineralogical and mechanical characterisation of coal mining wastes to obtain a concrete mix featuring strength and durability performance that meets specification. In the geotechnical literature, coal mining wastes are known to be highly heterogeneous and may host expandable minerals with potential durability problems. However, this review also found that simple geotechnical index tests can be conducted to yield useful information for the initial screening of coal mining wastes into a construction product. The state-dependent properties of coal mining wastes (e.g., water retention, hydraulic conductivity, shear strength) are found to be governed by complex factors such as coal content, particle size and shape, pore size and shape, and the presence and interaction of pore air and pore water in the void space, some of these are well-studied but much of these are to be further researched.

Keywords

Coal mining waste; Recycled geomaterials; Concrete; Construction

List of abbreviations

ACMW	activated coal mine waste
AMD	acid mine drainage
ASR	alkali-silica reaction
CMWG	coal mining waste geomaterial
CU, CD	consolidated undrained, consolidated drained
FI	flakiness index
GSD	grain size distribution
HREE	heavy rare earth element
LL	liquid limit
LREE	light rare earth element
OPC	ordinary Portland cement
m-CU, m-CD	multistage consolidated undrained, multistage consolidated drained
PL	plastic limit
PSD	pore size distribution
REE	rare earth element
SCM	supplementary cementitious material
SI	shape index
UC, UU	unconfined compression, unconsolidated undrained
XRD	X-ray diffraction

List of symbols

χ	effective stress parameter
ψ	pressure head
τ	shear strength
φ, φ'	total friction angle, effective friction angle
σ, σ'	total stress, effective stress
c, c'	total cohesion, effective cohesion
C_u	coefficient of uniformity
D_{50}	grain size for 50% finer by weight
D_s	fractal dimension of a grain size distribution
D_{s1}, D_{s2}	fractal dimension of populations 1, 2, respectively
d_s, $d_{s\ max}$, $d_{s\ min}$	grain size, maximum grain size, minimum grain size
$d_{s\ max1}$, $d_{s\ max2}$	maximum grain size of populations 1, 2, respectively
$d_{s\ min1}$, $d_{s\ min2}$	minimum grain size of populations 1, 2, respectively
Gs	specific gravity
Gs_{CMWG}	specific gravity of coal mining waste geomaterial
Gs_{coal}, Gs_{others}	specific gravity of coal and of materials other than coal
e, e_{max}, e_{min}	void ratio, maximum void ratio, minimum void ratio
$f(\ldots)$	function
I_d, $I_{d(2)}$	slake durability index, slake durability index of the second cycle
K, Ks	unsaturated hydraulic conductivity, saturated hydraulic conductivity
L	length
$M1$, $M2$	mass percentage of populations 1, 2, respectively
M_s	dry mass of particles
m_{coal}	coal content
q	flow rate
Sr	degree of saturation
s, s_e	matric suction, air entry/expulsion suction
u_a, u_w	pore air pressure, pore water pressure
z	length in vertical direction

1. Introduction

Coal mining waste geomaterials (CMWGs) consist of fragments of rocks and coal seams which are brought to the surface during coal extraction processes (Skarżyńska 1995a). Historically coal mining wastes have been viewed as problematic geomaterials. They are chemically heterogenous, prone to particle breakage by compaction, rapid degradation by wetting-drying cycles, and susceptible to liquefaction when loosely deposited. Furthermore, spontaneous combustion and leaching of acidic water to the surrounding environment are among the environmental challenges they present. These problems are associated with several special properties of coal and coal-bearing geomaterials in the wastes. Coal extraction is still ongoing, for example in 2019, the total coal production in Europe, North America and Asia Pacific amounted to 577.4 million tonnes (Mt), 701.5 Mt, and 5,911.8 Mt respectively (British Petroleum 2020). This ongoing production adds more CMWGs to the amount already in storage facilities (> 10,700 Mt by some estimates (Fan et al. 2014; Frías et al. 2012; Islam et al. 2021; National Research Council 2007; Skarżyńska 1995a; Zhao et al. 2008)) and imposes additional costs on producers and extra burdens on the environment.

In order to address the major challenges presented by coal mining in Europe and throughout the world, innovative concepts are being developed for managing, recycling and upcycling waste geomaterials generated by coal mining activities. One potential solution is to upgrade CMWGs as constituents of sustainable construction materials and products, and as such contributing to the establishment of a circular economy concept in the coal mining areas. In this respect there is a strong demand for geomaterials in the construction industry: for example 2.7 billion tonnes of natural aggregates are produced in Europe for construction purposes every year (European Aggregates Association 2017), meanwhile there is an imperative to conserve the natural resources. CMWGs have been utilised successfully in many low to medium level civil engineering applications such as controlled fills in mining zones, earthworks and land restoration, as rock armour in shoreline structures, aggregates in road construction and rail embankment (Hammond 1988; Skarżyńska 1995b). An enhanced understanding of the chemical and physical properties of CMWGs could accelerate their uptake and broaden their applications, particularly for higher level construction purposes.

To meet this aim, the properties of CMWGs need to be determined accurately, with a focus on their characterisation for reuse. Furthermore, the relationships between different properties and how some of them may be more relevant than others in specific engineering applications are key to investigate, together with some operational aspects of reclaiming the wastes from storage facilities.

Previous investigations into the (primarily geotechnical) properties of coal waste stockpiles and tailing lagoons were mainly driven by concerns about the stability of these structures. Compiled databases (Golder Associates Ltd 2015; ICOLD 2001; Rana et al. 2021) show that mine waste deposits pose a significant instability risk globally. Major failures of colliery tips and tailing dams are reported on a regular basis (Bishop 1973; Santamarina et al. 2017), some of most notable ones are listed in Table 1.

Table 1. Some failures of CMWG deposits investigated and reported in the literature

Time and location	Description	Potential causes of failures	References
21/10/1966, Aberfan, South Wales, UK	A flow slide involved approximately 107,000 m^3 of coal waste material.	The driving force of the failure was the buildup of pore water pressure at the toe of the tip, exacerbated by a loosely packed fabric of the fill making it susceptible to a flow liquefaction.	(Bishop et al. 1969)
26/02/1972, Logan County, West Virginia, USA	A flood involving approximately 498,000 m^3 of water was initiated by the failure of a coal waste embankment dam further upstream.	The upstream dam was made of coal spoils and its foundation consisted mostly of coal sludge. Piping in the foundation has probably led to excessive deformation and the subsequent overtopping of the dam.	(Davies et al. 1972)

1979-1991, Southwest Virginia, USA	11 slope failures in coal waste embankments required remediation.	Slaking and weathering of the embankment fill has probably reduced the shear strength to below what is required for stability. Failures were likely to have been initiated by the built-up of pore water pressure at the toe of the slopes.	(Donovan and Karfakis 2003)
2001-2002, Central Anatolia, Turkey	2 spoil pile instabilities involved more than 20 Mt of spoil material.	Gradual weathering and particle breakage by hauling, dumping and truck traffic have reduced the dominant grain size of the spoil material to silt-sized. Low residual strength was mobilised between the spoil and basal planar surface. Water pressure built-up from rainfall has initiated the instabilities.	(Kasmer et al. 2006)
Prior to 2004, Kalimantan, Indonesia	2 coal waste dump failures, the 1st involved 80 Mm^3 and the 2nd involved 10.5 Mm^3 of material	No cause of failure was stated, although it was emphasized that coal waste geomaterials were generally unsaturated, when water entered their pore space, they tended to slake, soften and weaken. A residual strength could have been developed along a thin layer, and the waste dumps could easily fail in a remolded mode.	(Pells 2016)
30/04/2004, South Field coal mine, Northern Greece	40 Mm^3 of dump materials was displaced up to 300 m from their original footprint, at a rate of 40-50 m/day.	The dump material was primarily low plasticity clays with local inclusions of silts and sand. The dump material has covered up a spring, choking its flow. Failure was possible due to a high-water pressure developing around the spring inside the spoil deposit.	(Steiakakis et al. 2009)
22/12/2008, Kingston Fossil Plant, Tennessee, USA	An uncontrolled release of 4.12 Mm^3 coal fly ash slurry was triggered by the rupture of a coal ash lagoon embankment	The embankment was built on a loosely-packed sluiced ash whose behaviour was contractive with a low undrained shear strength. Laboratory tests showed that the peak undrained shear strength of the sluiced ash was reached at 0.5% strain. At a higher strain, the undrained strength rapidly decreased to as low as 4.8 kPa.	(AECOM 2009), (TVA 2009)
31/10/2013, Alberta, Canada	A tailing pond embankment was breached, released 670,000 m^3 of coal waste water and 90,000 tonnes of fine particles into the Athabasca River	The embankment was overtopped due to a rise in the pond level prior to the full breach. Piping or retrogressive erosion of the upper loose material may have contributed to the initial overtop.	(HMTQ v. Prairie Mines & Royalty ULC 2017)

10/04/2020, Singrauli, India	400,000 tonnes of coal fly ash slurry were released from an impoundment facility, travelled a path of 6.5 km, spread an average width of 30m and an average depth of 1m.	The failure was triggered by an earthwork operator damaging a section of an embankment wall. However, the subsequent failure of the wall and the uncontrolled release of slurry was due to a severe hydrostatic pressure on the upstream of the embankment.	(Hiralal Bais v. Reliance Sasan Power Ltd. & Ors 2020)

There is a strong need to rehabilitate coal waste stockpiles and tailing lagoons due to their instability risk. However, any significant reclamation activity of coal waste from existing deposits should be carefully managed since reclamation of waste from a waste storage structure can alter its hydro-mechanical balance. The response which is triggered is dependent on mineralogical composition, particle and pore size distribution, fabric, water retention and permeability of the materials. The fabrics of coal waste stockpiles and tailing lagoons are also highly heterogeneous; the top few meters of a stockpile may be weathered while the materials at depths can be relatively fresh. Coal tailings are often stratified into layers of distinct particle sizes. Moreover, except those wastes buried permanently below the water table, most CMWGs are unsaturated, yet there are few experimental researches in the literature characterizing the unsaturated properties of CMWGs. The most recent studies (Fityus and Li 2006; Vidler et al. 2020) highlighted the importance of mineralogical composition, grain size distribution (GSD), pore size distribution (PSD), weathering and fabric, water retention and permeability. Characterising unsaturated properties of a CMWG is essential not only to understand its hydro-mechanical behavior and its use as compacted fill in geotechnical applications (Alonso and Cardoso 2010) but also for other foreseeable applications in the construction industry, including their use as constituents in concrete and cement-based materials, replacing, e.g., natural aggregates. Considering the substantial contribution of the construction industry to CO_2 emissions and natural resource depletion, such applications could hit the dual targets of serving the circular economy goals and minimising the adverse environmental impacts of both coal mining and concrete production. As such, significant savings could be made in the demand and use of natural raw materials for construction works, especially in areas near to active or decommissioned mines.

In order to turn CMWGs into valuable resources, particularly for construction, "fit-for-purpose" characterisations must be undertaken with regards to properties specific to their intended applications. Such characterisations will be detailed in this review paper. These properties include:

1. Hydro-chemical properties of repurposed CMWGs

A major concern associated with the reuse of CMWGs in the construction industry is the degradation/reaction of some of their constituents when they make contact with the hydrosphere (herein referred to as their hydro-chemical properties). This could potentially include generation of acid from the oxidation of disposed sulfide minerals when exposed to water or oxygen in the air (explained more fully in section 2.3.1), which would not only pose an environmental hazard but also influence the durability of the materials and (geo)-structures built using these materials. The determination of hydro-chemical properties of the repurposed CMWGs is necessary to determine and quantify the effects of key mechanisms responsible for ageing and degradation of the performance of repurposed CMWGs when exposed to various environments (e.g., salty, anaerobic, acidic, extreme climatic, etc.), and to understand their effects on the durability of the materials and products in which CMWGs may be incorporated. The current state of understanding the CMWG's hydrochemical properties is reviewed in section 3 of the paper.

2. Geotechnical index properties and hydro-mechanical behaviour of repurposed CMWGs

In order to provide a sound basis for the application of recycled CMWGs, it is important to evaluate their mechanical properties and to assess whether or not repurposed CMWGs can perform as well as natural geomaterials in various proposed "upcycling" applications. There are already clear evidences of recycled aggregates from the construction industry being used in new constructions (Guthrie and Mallett 1995; Silva et al. 2014) and, as such, by far most of the knowledge available in the literature is about the mechanical properties of recycled materials from the construction industry itself. Hence, properties of CMWGs relevant to these foreseen applications need to be reviewed and understood, also with reference to their outcome on the mechanical and durability performance of concrete and cement-based construction materials employing them as "secondary" raw materials.

The current state of understanding of CMWG's geotechnical index properties and hydro-mechanical behaviour is reviewed in section 4 of this paper. The focus will be on how some mineralogical compositions of CMWGs may be captured in geotechnical index properties and ultimately manifest in hydro-mechanical behaviours of CMWGs. It will be demonstrated, using data from the literature and original data from this study, that a geotechnical laboratory characterisation of CMWGs can be undertaken in a simple way, yet yield highly valuable information for the initial screening of CMWGs in specific applications.

2. Coal production and coal mining wastes

2.1. Coal production

Coal is mined by both underground- and surface-mining methods. Typical coal production and coal mining wastes are shown schematically in Figure 1. Depending on the amount and type of the coal (brown or black coal) available in an area, processing sophistication would differ hence producing different amounts and types of wastes (British Geological Survey 2010). Latest estimates put the current annual global coal production at 8,129.4 Mt (British Petroleum

2020). The total global coal reserve is at 1.14 trillion tons (EIA 2020), which is sufficient to last another 132 years at current rate of production.

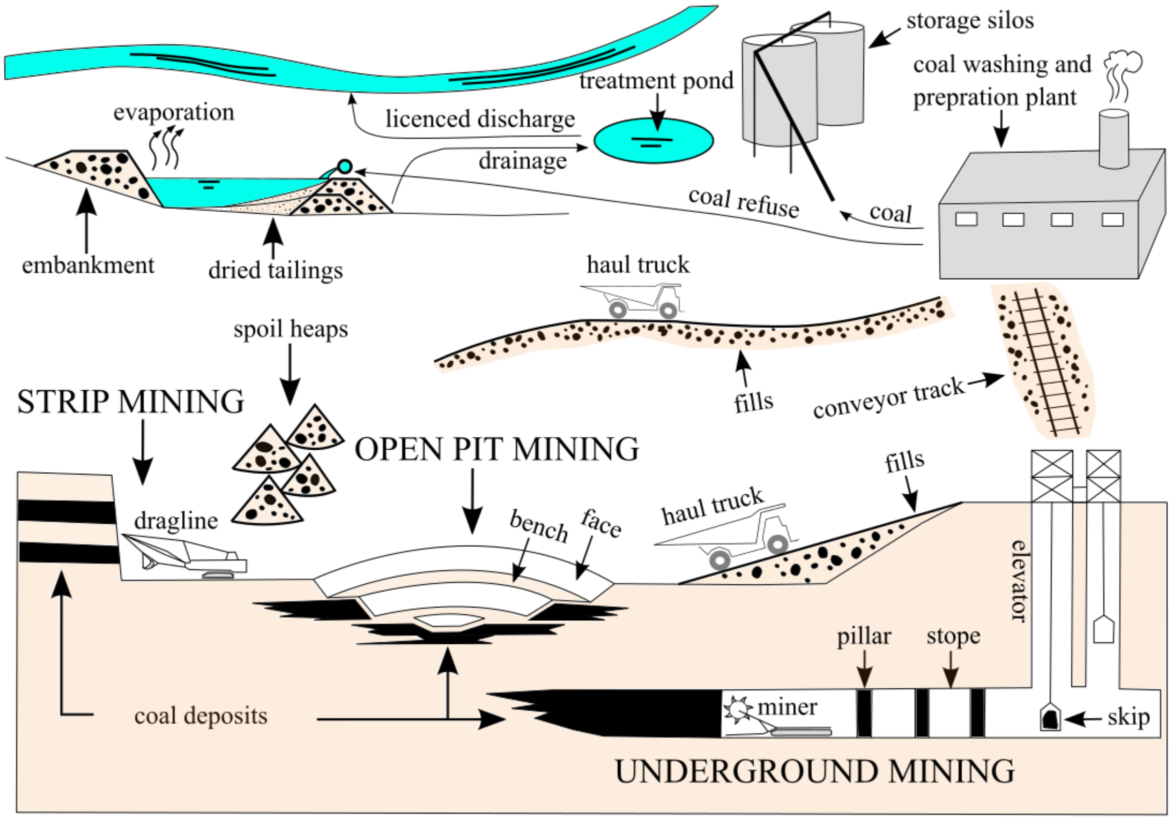

Figure 1: Coal production and coal mining wastes (adapted from British Geological Survey (2010), Coates and Yu (1977))

2.2. Coal mining wastes

Coal mining waste may be classified on the basis of its origin in a mine processing scheme: aggregate wastes include overburden and coarse rejects separated from the coal, and washery wastes comprise the finer fractions derived from the washing plant (Figure 2). Aggregate wastes and coarser washery wastes are typically disposed in spoil heaps, and finer washery wastes are disposed in slurry form in impoundment facilities. Figure 3 shows the coal mining wastes in a spoil heap from Forjas Santa Barbara (FSB) mine in Spain. Although coal mining wastes are increasingly being channeled into earthworks, roadworks, and are further processed into innovative construction materials (e.g., eco-efficient cements, brick and concrete blocks), there remains a significant amount generated by past and current mining operations that are directly dumped in spoil heaps and/or waste storage facilities. For example, in China, which is currently by far the world's single biggest producer and consumer of coal, about 36% of coal mining wastes are not utilised (Li and Wang 2019). There is, of course, a vast amount of historical coal mining wastes stored in spoil heaps and impoundments globally (> 6,600 Mt by Gutt and Nixon's estimate (1979)), and > 10,700 Mt by more recent regional estimates (Fan et al. 2014; Frías et al. 2012; Islam et al. 2021; National Research Council 2007; Skarżyńska 1995a; Zhao et al. 2008)).

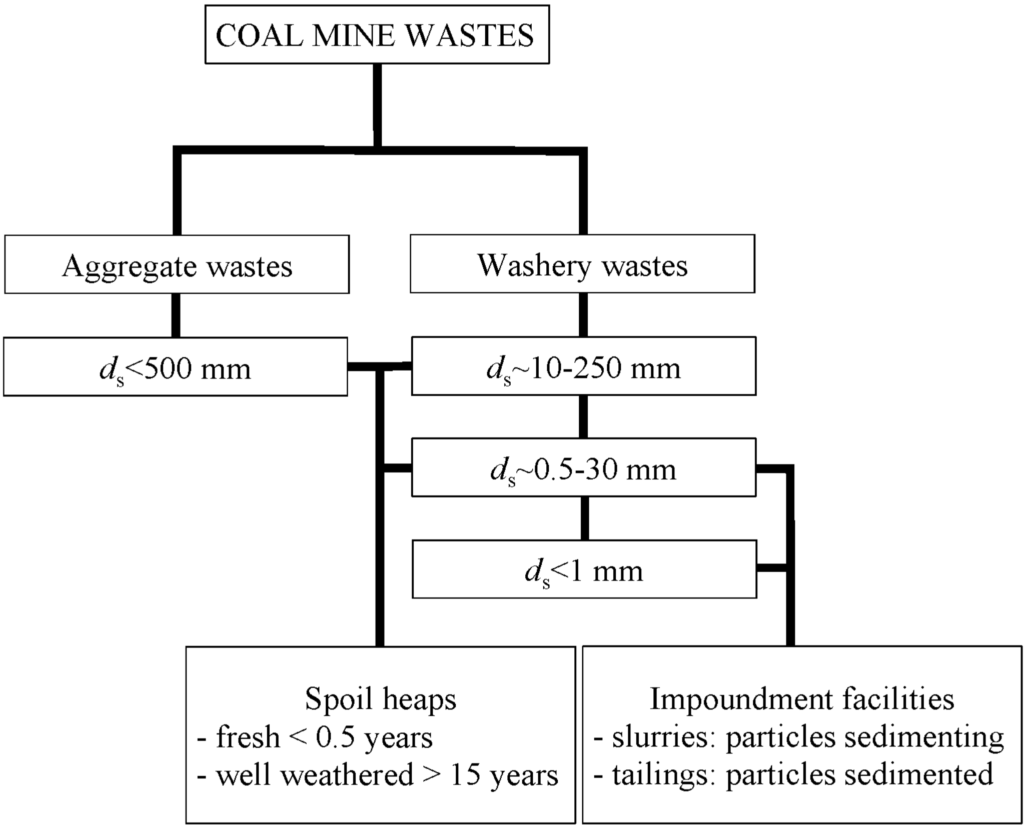

Figure 2: A classification of coal mining wastes (adapted from Skarżyńska (1995a)), d_s is grain size

Figure 3: Coal mining wastes in a spoil heap from Forjas Santa Barbara mine, Spain

Many CMWG deposits may be in metastable states. Past investigations (Table 1) show that failures of colliery spoil heaps were significantly contributed by shear strength degradation due to weathering, instability of a loosely packed fabric formed by dumping or poor compaction, combined with adverse structural features and a triggering event. Failures of coal tailing and ash ponds have been significantly contributed by their loosely packed fabric, unevenly placed slurry pumping, combined with adverse structural features and a triggering factor such as seismic excitation or heavy rainfall. Major failures of colliery tips and tailing dams (some with catastrophic consequences) are reported on a regular basis (Bishop 1973; Santamarina et al. 2017). There is a clear need to increase the effort currently put towards rehabilitating these CMWG deposits.

2.3. Hazards posed by coal mining waste

The high volume of residues generated by mining activities are usually put into storage facilities; the characteristics of the storage/disposal site being of paramount importance in order to handle the environmental hazards that may occur, as pointed out by Twardowska et al. (2004). The coal waste aggregates are usually simply stockpiled by the mine, compacted as fills or used in the base of tailings dam embankments (Figure 1). After a mine operation has concluded, it is a common practice to refill the excavation with the generated solid wastes. The environmental impacts of the mine will then depend mainly on how this stockpiled waste subsequently interacts with the atmosphere, rainwater, and in particular, groundwater.

The most well documented environmental impact of stockpiled coal waste is the seepage of acidic water: a phenomenon known as acid mine drainage (AMD). The acids generated may contaminate the nearby water sources and thus must be treated following disposal. Indeed, many aspects of stockpile design are intended to prevent/retard AMD, or capture the seepage water so it can be treated. In the refining plant the coal is ground and mixed with water, consequently generating a significant amount of slurry waste. On occasions, the contaminated slurry has been disposed in rivers or the ocean, or piped into storage facilities or underground. A common prevention strategy is to encapsulate wastes identified as potentially acid-generating within those that are acid-neutralising (by virtue to their carbonate-bearing mineralogy). The causes and effects of AMD are described in more detail in section 2.3.1.A, whereas possible bi-product of AMD, the combustion of coal waste stockpiles, is described in section 2.3.2.

A documentation of the major impacts produced by coal mining waste more generally has been developed by Younger (2004), who identified the following five items: air pollution, fire hazards, ground deformation, water pollution, and water re-source depletion. Disposal of coal mining waste, like any proposal for its reuse as a secondary raw material for civil engineering or construction, hence requires a robust understanding of the geochemical processes it is likely to undergo. Together with the physical and mechanical properties of the waste, which are summarized in section 4 of this paper, geochemical processes are explored in the reminder of this section and in section 3.

2.3.1. Acid Mine Drainage

CMWGs often contain minerals that are chemically unstable in the presence of oxygen and water, which may dissolve after prolonged exposure to rainwater/groundwater. The most common such minerals are the sulfides (chiefly pyrite and pyrrhotite), and their dissolution can substantially lower the pH of the contact water. This phenomenon is termed as acid mine drainage (AMD), and is well documented at many coal mines around the world (INAP 2009). AMD is a major environmental concern because in addition to the ecological hazard presented by low pH, such acidic water has an enhanced capacity for dissolving toxic metals that are frequently present as trace elements in coal, such as lead, arsenic, cadmium, selenium, copper and zinc (Park et al. 2019). AMD therefore has the potential to widely disperse toxic metals, contaminating major aquifers and watercourses far beyond the location of the source materials. In the construction industry, the principal hazards posed by sulfide oxidation are the expansive stresses generated within concrete when iron- and sulfate-bearing compounds such as ferrihydrite or gypsum precipitate from acidified porewater (Chinchon et al. 1995).

Although there are many chemically reduced species that can contribute to AMDs, sulfide is overwhelmingly the most common species found in CMWGs (INAP 2009). Among the sulfide-bearing minerals pyrite (FeS_2) is the most abundant (Nordstrom 2011, Park et al. 2019), and its net oxidation to yield sulfuric acid can be written as follows:

$$\mathrm{FeS_2} + \frac{15}{4}\mathrm{O_2} + \frac{7}{2}\mathrm{H_2O} = \mathrm{Fe(OH)_3} + 2\,\mathrm{H_2SO_4} \qquad (1)$$

Whilst the expression in Eq. (1) usefully indicates the 1:2 molar ratio between the amount of pyrite oxidized and the amount of acid ultimately generated, it does not describe the oxidation process comprehensively. In particular, the oxidant in the reaction need not be molecular oxygen, and at pH lower than around 3.5 ferric iron (Fe^{3+}) can assume this role if it is available:

$$\mathrm{FeS_{2(s)}} + 14\mathrm{Fe^{3+}} + 8\mathrm{H_2O} = 15\mathrm{Fe^{2+}} + 2\mathrm{SO_4^{2-}} + 16\mathrm{H^+_{(aq)}} \qquad (2)$$

This alternative oxidation pathway is often sustained by microorganisms which maintain the supply of ferric iron by oxidizing ferrous ion (Fe^{2+}) (Crundwell 2003). A decrease in pH caused by the oxygen-mediated reaction (the first equation above) can create the conditions necessary for this alternative Fe^{3+}-driven process, which can proceed 2 to 3 orders of magnitude faster (INAP 2009). The oxidation rate may however be reduced by the presence of neutralizing materials such as calcite, which can maintain near-neutral pH if suitably distributed within the waste. A further important influence on reaction rate is the surface area of the pyrite, which can be orders of magnitude larger for intricate ‘framboidal’ growths than for the simpler ‘euhedral’ crystals.

In summary, the rate of sulfide oxidation within a particular CMWG is a complex function of the dissolved oxygen concentration and the pH of the contact water, the exposed surface area of the sulfides, and the chemistry of the accompanying minerals.

2.3.2. Combustion

Although CMWGs are materials that have been rejected from the coal production process, this does not preclude them from containing percentage-level concentrations (by mass) of carbonaceous material. This coal can ignite if not stored or transported appropriately (Onifade and Genc 2020). Spontaneous combustion is commonly observed at coal mines in coal spoils (though less commonly in overburden materials) and is often initiated by sulfide oxidation (see section 3.1.2.3), which is a strongly exothermic process. Spontaneous combustion at mine sites is generally mitigated by limiting the thickness of waste dumps (to promote cooling), constructing them in wind shadows where possible (to minimize headwind-assisted oxidation), and placing lower limits on material grain-size. The application of CMWGs as aggregates will require similar considerations, especially if the conditions envisaged are likely to permit sulfide oxidation (e.g., continuous exposure to moist air).

3. Upgrading CMWGs as raw materials in construction industry

The mineralogy of CMWGs varies widely, since it depends on the mineralogy of the geological formations overlying or adjacent to the coal seam being mined. Consequently, no single shortlist of minerals can be said to comprise CMWGs exhaustively, although the range of possibilities is restricted by the sedimentary origin of the coal, which typically requires the adjacent 'host' formations to also be sedimentary. Exceptions include certain types of sediment which contain fragments of ancient igneous rocks ('breccias'), and mines at which some of the rocks overlying the coal seam (the 'overburden') are not of sedimentary origin.

Consequently, CMWGs usually comprise of minerals found in the sedimentary rocks; minerals that are rich in silicon and aluminum oxides, chemically stable at near-surface conditions, and frequently hydrated. These include quartz, varieties of clay (e.g., kaolinite and illite), potassium feldspar, micas, carbonates (calcite, dolomite), and less commonly sulfates, halides, iron oxides and amphiboles. The nature of coal formation, which invariably comprises the accumulation of plant matter in a stagnant, often water saturated environment, followed by diagenesis, dictates that certain similar mineral transformations have been documented to occur across different coal deposits world-wide. For example, coal seams of the Permian Shanxi formation in Juye coalfield, China, are dominated by kaolinite and calcite, with claystone and sandstone interburden dominated by kaolinite, montmorillonite, and illite; and quartz, chalcedony, feldspar and kaolinite respectively (Zhang et al. 2019). These minerals are derived from volcanic and granitic source rocks in the pre-diagenetic period which then underwent weathering in a peat swamp environment; such acidic conditions are favourable for the weathering of feldspar and formation of kaolinite (Zhang et al. 2019).

Besides these ‘major’ constituents, a wide range of minor constituents have been reported for CMWGs. Again, such constituents can be highly variable, however, the nature of coal formation, and its resultant physical and chemical properties dictate that certain minor constituents commonly exist. For example, sulfide bearing minerals, such as pyrite and chalcopyrite also form under oxygen-poor conditions and are therefore common in CMGWs. Such minerals, in addition to coal, are typically reported as most relevant for CMWG reuse potential due to their capacity to inhibit the geotechnical performance of CMWG-bearing construction materials (Section 3.1.2). Another common constituent of coal-bearing strata is the radioactive and ecotoxic element uranium. This co-association is due to the fact that carbon-rich organic matter within the coal-bearing strata can act as a potent sorbent and reducing agent, which when exposed to groundwater containing uranyl ions, can result in their selective precipitation as solid (and surface bound) mineral phases. Such occurrences are widespread; for example, Huang et al. (2012) undertook a literature survey of uranium occurrence in 1184

Chinese coal samples with a range typically within 0.5-10 mg/kg, and an average of approximately 3 mg/kg.

Despite such common constituents the typically diverse mineralogy of CMWGs gives rise to a similarly wide range of geotechnical and geochemical properties. As such, the suitability of CMWGs for a particular construction application can vary from one mine to another. While the presence of certain minerals is desirable for some applications (aggregates require hard minerals like quartz for example), as mentioned earlier, the absence of others can be just as important (such as sulfides in concrete additives). The effects of mineralogy on the main applications of CMWGs are discussed in the following subsections, with a focus on chemical processes than geotechnical properties (which are discussed in section 4).

3.1. Chemical properties necessary for use of CMWGs as raw materials in construction industry

CMWGs have been investigated as a replacement for both aggregates and binders (cements) in concrete and cementitious composites. The re-purposing as construction aggregates is reasonably established for low-grade applications such as road-fills (Amrani et al. 2020), reclamation fills (Indraratna et al. 1994; Rujikiatkamjorn et al. 2013), road/railway embankments (Wilmoth and Scott 1979) and in various other applications where limited amounts of Portland cement are used for stabilization and performance improvement (González Cañibano 1995; Okagbue and Ochulor 2007).

Use as concrete aggregate is a significantly more lucrative application because the specifications are more demanding; not only must the CMWG’s constituent minerals have sufficient strength to meet the geotechnical specifications to be employed as concrete aggregates, but they must not undergo adverse chemical reactions with the cement (i.e., they must be chemically stable). CMWGs have been used successfully to manufacture fine aggregates for some low-spec concretes such as paving blocks (Rossi dos Santos et al., 2013), whereby the waste fraction with relative density/specific gravity 2.4-2.8 was separated, crushed, and substituted for the conventional aggregate (sand) in varying proportions. Compressive strength and abrasion resistance tests indicated the maximum acceptable substitution to be 50% by volume. Modarres et al. (2018) investigated the use of different types of CMWGs, including powder, ash and aggregates, in roller compacted concrete for pavements, in replacement percentages up to 20% by volume, demonstrating that in all cases (but for the 20% powder replacement) the required specifications (about 28 MPa cube compressive strength at 28 days) were successfully met. Best results, also in terms of toughness, were obtained with relatively low replacement percentages (5% by volume), irrespective of the CMWG type, with levels of performance comparable to that of the parent mix, if not slightly better, due to the filler effects of the CMWGs. Higher replacement percentages resulted in performance deteriorating.

Wang and Zhao (2015) produced a series of concretes using Chinese coal gangue as coarse and fine aggregate to determine the influence of gangue grading on their geotechnical properties. Fuller’s curve n values ranged from 0.44 to 0.68, and 0.62 was identified as the optimal one, resulting into a maximum 28-day compressive strength of the concrete equal to 37MPa. Li et al. (2021) investigated the microstructural and geotechnical properties of concrete prepared using a coal gangue as aggregate, dominantly composed of silica, kaolinite and calcium carbonate. The concretes obtained were significantly weaker than those prepared using quartz gravel as aggregate.

3.1.1. Pozzolanic activity

A desirable potential application for CMWGs is their utilization as a supplementary cementitious material (SCM) in blended concrete. Such blends are increasing in popularity as the construction industry seeks to reduce its carbon emissions by finding supplements for cement, whose production is extremely energy and carbon intensive (4-5 GJ and ~800 kg emission of CO_2 per ton of ordinary Portland cement (Shamir et al. 2020)). CMWGs are good SCM candidates because they often contain large fractions of clay minerals that can be conditioned to acquire pozzolanic properties. These clays, kaolinite in particular, must be thermally activated by calcining between 500 and 900 °C for around 2 hours (Frías et al. 2012; Vigil de la Villa et al. 2014), the precise conditions are somewhat varying for the different CMWGs tested to date. This treatment dehydrates kaolinite to form metakaolin, a semicrystalline compound whose pozzolanic activity arises from its propensity to react with portlandite ($Ca(OH)_2$) in cement to form cementitious calcium silicates (Bich et al. 2009). Mixtures of such thermally activated wastes with conventional cement, ground to particle sizes of order ~75µm, have yielded concretes with properties comparable to those employing purely conventional cement.

Much of the research into CMWGs as pozzolans to date has been conducted on Spanish waste materials. Thermally activated CMWGs from Spain have been repeatedly used to replace between 20% and 50% of ordinary Portland cement (OPC), yielding concretes with tensile strengths and corrosion resistance comparable with those containing OPC exclusively. Such products have been manufactured using both coal aggregate waste (Caneda-Martínez et al. 2019; García Giménez et al. 2016; Vigil de la Villa et al. 2014) and coal washery waste (Frías et al. 2012; Rodríguez et al. 2021). For example, Frías et al. (2012) synthesized concrete blends containing (individually) coal washery waste and coal aggregate waste as substitutes for up to 20% of the OPC. Their products were type II/A cement compliant with European standards with respect to sulfate and chloride concentration, as well as meeting European setting time, volume stability and strength requirements. Caneda-Martínez et al. (2019) found that the corrosion resistance of rebar to chloride ions was improved by the substitution of 20% of the

OPC by thermally activated CMWGs. In general, these CMWG-blends require higher water/cement ratios than conventional OPC, e.g., approximately 13% more in blends employing 20% SCMs (Frías et al. 2012).

Similar results were obtained by Vegas et al. (2015) who studied the performance of blended cements with CMWG replacements of up to 20%. Small to moderate replacement percentages (6% to 10%) led to slight increase of compressive and flexural strength in the short term and a moderate decrease in the longer term (> 90 days), whereas a slight decrease in strength (less than 10%) was always observed for the highest investigated replacement percentage. Significantly, drying shrinkage was also increased by the replacement of OPC with CMWGs. This has been attributed to the following phenomena: CMWGs accelerating the hydration of cement; pozzolanic reactions between the metakaolin contained in the CMWGs and calcium hydroxide from the hydration of cement clinker; and lastly, to an increase in capillary pressure that is a consequence of a change in the pore size distribution.

While studies on CMWGs to date have focused on metakaolin as the source of pozzolanic activity, it is not necessarily the only such constituent in the waste media. Kaolinite concentrations in the CMWGs investigated varies widely, from about 70% to as low as 14% (Rodríguez et al. 2021), suggesting that kaolinite might not be the only constituent that yields pozzolanic properties upon thermal activation. The possibility of alternatives is well demonstrated with the success observed by employing coal fly ash as a pozzolanic replacement (Jovanovic et al. 2014; Shamir et al. 2020), which mainly comprises aluminosilicate glass (the first known pozzolans, sourced from southwest Italy, which were also glassy in nature) rather than metakaolin or any other clay or clay derivative. Another alternative pozzolan synthesis method has been demonstrated by Wang et al. (2021), who combined sodium hydroxide solutions with various mixtures of coal gangue and blast-furnace slag to yield prototype road-stabilization materials (a less demanding application, but nonetheless a legitimate example of pozzolanic activity).

We venture here to suggest that thermal activation of CMWGs could remove their constituent sulfides by oxidizing them directly to SO_2 gas and hematite. Hu et al. (2006) reviewed studies of pyrite oxidation in air and reported that this decomposition reaction occurs at less than 800 K; a similar temperature to that employed for thermal activation of kaolinite. The possibility that a single such thermal treatment (or some specially optimized variant) might both produce pozzolans and suppress undesirable sulfide oxidation (see section 3.1.2.3) is worthy of further investigation.

3.1.2. Chemical processes deleterious to use of CMWGs as construction materials

3.1.2.1.Alkali-Silica Reaction

The presence of amorphous silica in concrete aggregate can cause swelling and spalling (Figure 4) if it reacts with hydroxide compounds within the cement; a phenomenon known as alkali-silica reaction (ASR). The problematic reaction products are calcium silicate hydrate gels, which are hygroscopic and generate large internal stresses if they absorb water from the concrete pore solution (Fanijo et al. 2021). CMWGs may contain many varieties of amorphous silica (such as chert or opal) or strained quartz (which is also vulnerable to ASR), since these are also common in sedimentary rocks. Thorough petrographic inspection of CMWGs to search for these amorphous phases is essential if they are to be used as concrete aggregates. Inspection procedures appropriate for this task have been standardized (e.g., ASTM C1567-21, ASTM C1260-21) which include experimental tests as well as petrographic examination. It should be noted that to the authors knowledge, to date, ASR has not been reported in concretes prepared using CMWGs as aggregate. Since this application remains relatively untested, and since ASR is a chronic condition that can take years to develop, little can be inferred about the propensity for CMWGs in general to promote this reaction. Interestingly, the use of SCMs, including coal fly ash, has been reported to mitigate damages from ASR (e.g., Shafaatian et al. 2013), although the mechanism responsible is not known with certainty. Indeed, the mitigation of ASR has become a motivation for using SCMs, underlining the interdependence of the different chemical processes described in this review.

Figure 4: A highway barrier deformed by alkali-silica reaction (ASR) (Akhnoukh 2013).

3.1.2.2.Alkali-Carbonate Reaction

The alkali-carbonate reaction (ACR) is a deformation-inducing chemical process analogous to the ASR, in which carbonates rather than amorphous silica reacts with hydroxide from the cement to form a hygroscopic gel. The degree to which carbonate decomposition (known as dedolomitization) is responsible for deformation is uncertain however, since carbonates are popular concrete aggregates and yet most do not exhibit ACR (Aquino et al. 2010). For example, Katayama (2009) and Grattan-Bellew et al. (2010) have suggested that some cases of ACR are actually instances of ASR, where the role played by the carbonates is to contribute amorphous silica (silicious dolomites are common geological materials); the dedolomitization itself being merely incidental. Since calcite and dolomite are both common constituents of CMWGs, the presence, abundance, and chemistry of these minerals should be carefully determined in any CMWG before it is considered as a concrete aggregate.

3.1.2.3.Sulfate attack

Sulfide oxidation has been documented in concretes that contain sulfide-bearing aggregates, and typically manifests as yellow discolouration accompanying a distinctive network of cracks (known as map cracking) and pits/voids (known as pop-outs). These fractures critically undermine the strength of the concrete, sometimes necessitating preemptive demolition of the building. The onset of such damage can be rapid and appearing after as early as 3 years (Rodrigues et al. 2012). In the concrete industry, this propensity for sulfate ions to react with components in cement is known as sulfate attack (Müllauer et al. 2013), and the expansive precipitates responsible for the fractures vary, depending on the chemistry of the aggregate and cement. Importantly, sulfate attack can occur when the sulfate source is external to the concrete, such as from contaminated groundwater or sewage. In cases where the source is endogenous, oxidation of both pyrite and pyrrhotite have been observed (Schmidt et al. 2011). In response to the recognition of this phenomenon, regulations have been introduced that specify the maximum abundance of sulfide in concrete aggregates. These differ by country, but most agree that it should not exceed 1% by mass, and must exclude framboidal pyrite crystals potentially present in CMWGs in problematic quantity (see section 4.2.2).

3.1.2.4.Chloride induced corrosion of steel reinforcement

Caneda-Martínez et al. (2019) studied how the presence of activated coal mine waste (ACMW) in concrete affects the corrosion of steel related to the chloride ion concentration of the concrete porewater. When a certain threshold of chloride ion content is reached, the protective layer around the steel bars, created by the highly alkaline cement pore solution, is destroyed, which makes them more susceptible to corrosion. A chloride-induced accelerated corrosion test was conducted on steel bars embedded in four different mortar specimens: a reference sample and three others with partial substitution of OPC by activated coal mining waste (substitutions of 10%, 20% and 50% by volume). It was concluded that the addition of ACMW to concrete induces a decline in critical chloride ion content by up to 90% when compared to the reference specimens (i.e., it made the steel more susceptible to corrosion). On the other hand; however, it was also found that mixes with CMWGs had a longer corrosion onset time, due to higher resistance to chloride ion penetration and lower chloride diffusion coefficients, most likely promoted by the pozzolanic activity of the CMWGs (see section 3.1.1).

3.1.3. Required chemical properties of CMWGs to be used as raw materials in construction industry

Because of the aforementioned deleterious processes, the European standard EN 1744 prevents the use of reactive aggregates, as per alkali silica reaction, complying with expansion limits measured according to a suitable accelerated test. The same standard also limits the chloride content of aggregates to 0.03% by mass and that of sulfates to 0.2% and 0.8% for coarse and fine fractions respectively. The total content of sulfur, which may also be present into other compounds, shall not exceed 1% by mass of aggregates (2% for blast furnace slags).

Presence of other substances, especially organic, which may affect the setting time of the concrete is limited to amounts which would not increase the setting time by more than 120 minutes and would not cause a reduction of the compressive strength at 28 days by more than 20%.

4. Relationship between mineralogical and geotechnical properties of CMWGs and the required characteristics as recycled aggregates.

4.1. Required physical and mechanical properties of CMWGs to be used as secondary raw constituents in construction materials

According to the European standard EN12620:2002+A1:2008 aggregates are the granular materials used in concrete batching and may be natural, manufactured or recycled. Recycled aggregates are classified according to their origins into concrete and concrete products, including concrete masonry units; unbound and hydraulically bound aggregates; masonry units made of clay, calcium silicate or aerated concrete blocks; bituminous materials; glass; floating material and miscellaneous, including metals, non-floating woods, rubber and plastic and soils. Depending on their origin, the maximum percentages of constituents in the recycled aggregate fractions are defined.

Determination of geometrical properties of aggregates is governed by EN 933 standards (including 11 different parts). Besides the grain size distribution that is necessary to sort the aggregates in concrete according to the best grading curve, for the use of CMWGs as aggregates in concrete and construction industry the following properties are of interest:

- the flakiness index (FI), defined as the percentage, by weight, of particles in an aggregate which have their average least dimension (thickness) less than 0.6 times their average dimension;
- and the shape index (SI), defined as the percentage, in mass, of the non-cubic particles present in the test portion are also defined to be met by any material to be used as aggregates.

Determination of physical and mechanical properties of aggregates is on its hand governed by EN 1097 standards (10 parts). Compressive strength and resistance to wear and fragmentation are the most relevant mechanical properties to be measured, whereas for the physical ones, bulk and grain density and water absorption are of paramount interest, the latter being correlated also to freeze and thaw resistance of the aggregates and hence of the concrete as well.

There are fundamental reasons for the standards' requirements on the physical and mechanical properties of CMWGs to be used as secondary raw constituents in construction materials. Sections 4.2 and 4.3 present an updated review of studies on these rationales at the scales of intact coal mining aggregates and assemblies of coal mining aggregates.

4.2. Mineralogical and index properties of CMWGs

4.2.1. Influence of coal content on the specific gravity (G_s) of CMWGs

The percentage of coal in a CMWG deposit is influential to many of its properties, including specific gravity (G_s). The G_s of coal is reportedly between 1.27-1.47 (Nebel 1916; Skarżyńska 1995a). Depending on the amount and the type of geomaterials co-existing with coal in the CMWGs, G_s of aggregate waste is estimated to be in the range of 2.3-2.5 (Skarżyńska 1995a), and G_s of washery waste is smaller, at 1.75-2.15 (Leventhal and de Ambrosis 1985), reflecting the additional coal extracted by the washery process. Following the general definition of G_s (Kirby 1980), it can be shown that

$$G_{s_{CMWG}} = \frac{1}{\left(\frac{1}{G_{s_{coal}}} - \frac{1}{G_{s_{others}}}\right) m_{coal} + \frac{1}{G_{s_{others}}}} \qquad (3)$$

where m_{coal} is the percentage of coal in mining wastes by dry mass. This relationship is plotted on Figure 5 assuming $G_{s_{others}} = 2.65$ together with data of Kirby (1980) and Yasser et al.

(2004). Also plotted in Figure 5 are data from this study for samples obtained from 6 Polish mine sites. The G_s of a CMWG could be determined more easily than its carbon content in the laboratory. A drying oven, a volume measuring device and a balance are sufficient to determine G_s to a reasonable accuracy (+/- 1 decimal point). Knowing the G_s of a CMWG, Figure 5 could be used to approximate the corresponding carbon content of the CMWG.

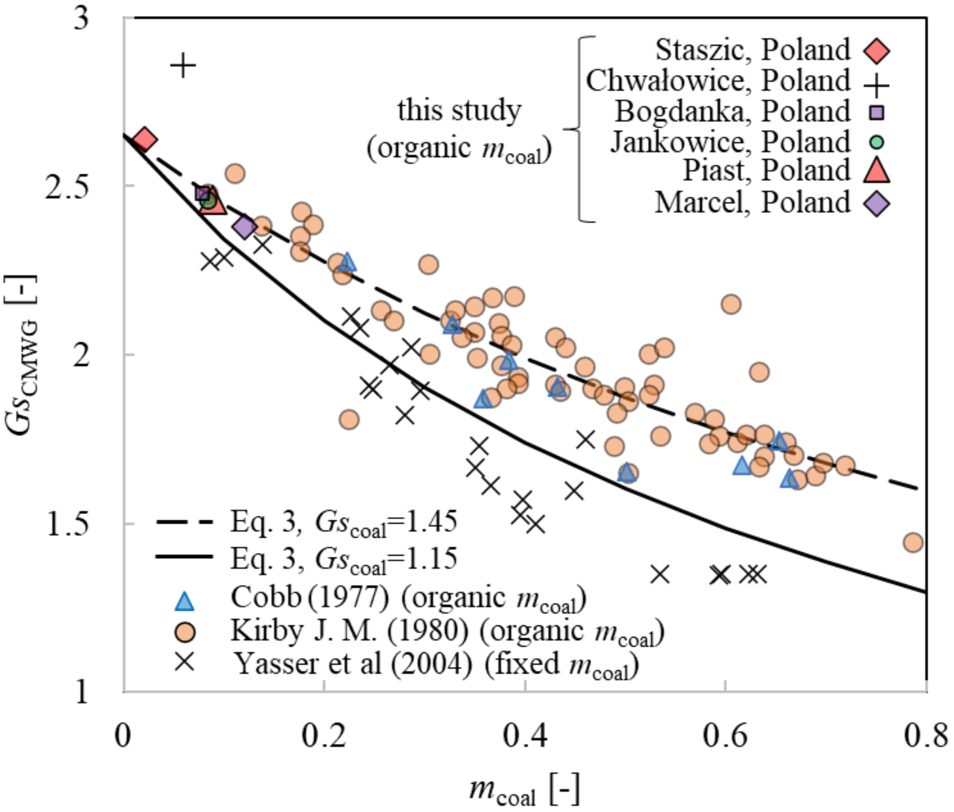

Figure 5: G_s of CMWGs correlated with their carbon content

4.2.2. Influence of expandable minerals on the durability of CMWGs and construction products containing CMWGs

The physical weathering of CMWGs is mainly driven by the presence of expandable minerals, in particular, montmorillonite and pyrite in their composition (Taylor 1974a; Taylor and Spears 1970). The expansion is greatest when sodium is present as interlayer cation (Mielenz and King 1952). Taylor (1974a) cited the example of a mudstone in the UK (i.e., Stafford tonstein) containing a high percentage of mica-montmorillonite and exchangeable Na+ that it disintegrates quickly and even completely when immersed in water. The primary mechanism that causes this type of disintegration is by way of air breakage or slaking (Terzaghi and Peck 1948). When initially-dried argillaceous rocks are wetted, water gradually fills void spaces and drives an increase in pore air pressure according to Boyle's law. The inflated pore air pressure causes the argillaceous rocks to fail along their plane of weakness, which for most sedimentary rocks, would be their bedding plane (Nezhad et al. 2018; Bagheri and Rezania 2021). Also, when rocks mined at depth are subsequently dumped in spoil heaps, the changeover from a high to low effective confining stress regime induces a volumetric dilation and accelerate their degradation. Another mechanism is due to a changeover from one environment to another, e.g., CMWGs originated from a saline environment when placed in contact with fresh water would be subjected to significant osmotic swelling pressure (Seedsman 1986).

When aggregates used in concrete contain significant amount of expandable minerals, there were many reports of subsequent deleterious volume change during wetting-drying cycles (Knight 1949; Rhoades and Mielenz 1948). Cemented soils with significant amount of expandable minerals were commonly observed to have major cracks attributed to drying-wetting cycles rather than external loads (Croft 1967). Byrd (1980) reported that the Canterbury bypass in the UK was constructed using CMWGs in sub-base layer but following a period of heavy rainfall, serious moisture swelling was observed at construction joints. Thomas et al. (1989) reported the results of a site investigation for three failed pavements in the UK. The pavements were built with cemented-stabilised CMWGs which apparently met the strength and durability requirements at the time of construction. Cored samples were collected from the three sites and subjected to a range of tests in the laboratory (i.e., compressive strength test, total-, pyritic- and sulphate-sulphur content test, X-ray diffraction, thin section examination, and scanning electron microscopy examination). They concluded that oxidation of pyrite mineral in the CMWGs had caused expansion of the cement-stabilised CMWGs and may have caused and/or exacerbated cracks in the pavements rendering them unserviceable.

Taylor and Spears (1970) divided clay and clay-associated minerals into three groups, a similar division was adopted: kaolinites and minor chlorite, illite and muscovite, and expandables (montmorillonite, vermiculite, mix-layers, pyrite, calcite). The compositions of those minerals in some CMWGs reported in more recent literature are shown on Figure 6. Also plotted is data from this study related to the FSB mine in Spain.

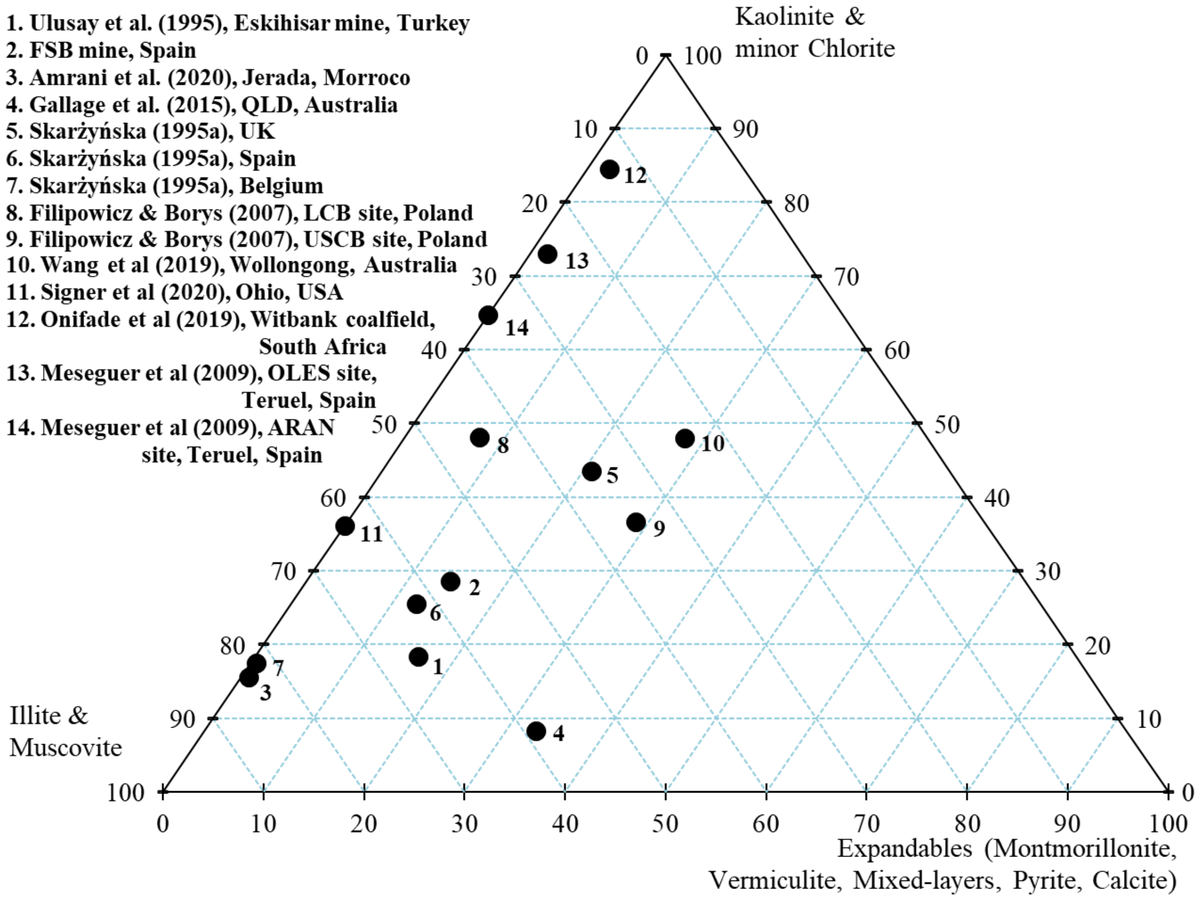

Figure 6: Composition of clay minerals in some CMWGs reported in the literature

Attempts have been made to quantify the propensity of argillaceous rocks to slake using mechanical tests. Among them, Franklin and Chandra (1972) developed the slake durability test to evaluate the potential of shales, mudstones, siltstones and other clay-bearing rocks to resist the weakening and disintegration resulting from drying-wetting cycles. In essence, a mass of dried rock is rotated inside a perforated drum half-immersed in a water bath at 20°C for 10 minutes. A slake durability index I_d is calculated as the percentage ratio of the final to initial dry weights of the rock in the drum i.e., the higher I_d is, the more durable it is. The test has been standardised (ASTM D4644-04 (2004), ISRM (1979)) where two drying-wetting cycles are specified and the slake durability index of the second cycle $I_{d(2)}$ is reported.

Adaptations to the slake durability test have been made, given the wide variety of mineral compositions and environments argillaceous rocks are exposed to. Gökçeuğlu et al. (2000) collected 141 samples of weak and clay-bearing rocks from different parts of Turkey and subjected them to four drying-wetting cycles of slake durability tests, XRD and uniaxial compression tests. They found that the durability of clay-bearing rocks correlates best with the amount of expandable clay minerals. They conducted a statistical analysis to show that strength probably has no influence on the durability of laminated marls (there might be an association). Miščević and Vlastelica (2011) conducted the cycle slake durability test to characterize marls from Dalmatia in Croatia. They adopted four drying-wetting cycles because they argued that by the end of the 2nd cycle, although many lumps of marl did not pass through the openings of the drum, the rock itself had practically disintegrated. They performed accompanying strength tests and concluded that strength probably has no influence on the durability of the marls. In another notable study, Vallejo (2011) conducted point load tests, slake durability tests and thin section examinations of 68 shale samples from the Appalachian region of the United States. They found that pore micro-geometry has a major influence on the degradation of the shales, in that the air breakage mechanism was more effective in causing the slaking of those shales with smooth pore boundaries than those with rough pore boundaries. Qi et al. (2015) conducted a static slake durability test involving 10 wetting-drying cycles on a red mudstone taken from a depth of 154.10–162.05 m in a coal mine in Shandong (China). They found that as the slaking progresses, the number and size of pores and fractures increase, the structure of the surface of the slaked particles becomes more disordered and complex. They also reported that when the particle size of the stone is reduced by slaking to below 5 mm, it becomes more durable.

Some durability testing has also been conducted on stabilised geomaterials. Surendra et al. (1981) studied how additives might be added to nondurable shales to improve its performance during their placement and service as an embankment using the slake durability test. They reported that adding lime (up to 7% of a rock's dry mass) to Osgood shale showed little improvement while adding it to New Providence shale showed a substantial improvement. The shales themselves were similarly nondurable but contained very different exchangeable solution percentages. Kettle (1983) conducted laboratory and field trials on 10 CMWGs collected from major coal fields in the UK. The CMWG samples were screened for their compliance with UK requirements at the time (Department of Transport 1977), among which were LL<45% and PL<20% and the coefficient of uniformity C_u<5. They were either untreated (in which case, they were prepared as samples and tested immediately) or stabilised with cement at 5%, 10% of their dry mass, cured for 7 days at 20°C and atmospheric confining pressure, then subjected to a range of strength tests, frost heave test (Croney and Jacobs 1970) and immersion test (BS1924 1975). It was found that some CMWGs could be cement-stabilised to function satisfactorily as road subbase and base materials. However, frost heave and immersion tests showed that those CMWGs with significant fines (>30% finer than 75 μm) were not sufficiently durable. Stavridakis and Hatzigogos (1999) created clayey admixtures in the laboratory containing between 0% and 45% montmorillonite (in terms of dry mass), the others being sand and kaolinite. They stabilised the mix with 4% and 12% cement then conducted standard slake durability tests on the hydrated material. They found that the admixtures with a liquid limit of 40% can be treated satisfactorily with 4% cement (in that it is sufficiently durable for its purpose). Although the admixtures with a liquid limit of 60% can be stabilised satisfactorily with 12% cement but that was considered uneconomical. In a recent study, Liu et al. (2020) conducted three wetting-drying cycles slake durability tests on a paste backfill comprising cement, fly ash and sand mixed according to a recipe. Their results showed that a lower hydraulic conductivity contributed to a more durable paste back fill material. Their microscopic analysis showed that the durability of the material might be linked to a non-uniformly distributed pore structures although the mechanisms for this remain unexplored.

4.2.3. Influence of clay minerals on the plasticity of CMWGs and construction products containing them

The plasticity of geomaterials can be attributed to the presence of clay minerals in their make-up (Rezania et al. 2020). The liquid limit (LL) and plastic limit (PL) are the water content at which a CMWG starts to flow like a liquid and the water content at which a CMWG transits from brittle to plastic deformation, respectively. LL is determined by Casagrande's percussion method or the fall cone method, and PL by the rolling thread method. The plasticity index, I_p, is determined as LL-PL. The mechanisms that enable brittle failure in the plastic limit test are by air entry or cavitation (Bagheri et al. 2018; Haigh et al. 2013; Sivakumar et al. 2009; Vardanega and Haigh 2014), and the governing factors are complex: mineralogy, structure, texture, etc., with mineralogy playing a key role (Fleureau et al. 2002; Williams et al. 1983). The presence of even a small amount of clay minerals can impact engineering behaviors of a geomaterial significantly, thus LL and PL feature in the unified soil classification system for classifying fine-grained geomaterials, and coarse-grained geomaterials with significant fines. CMWGs may be sorted into different sizes when used as aggregates in construction products. The suitability of fine-grained CMWGs as recycled materials is strongly dependent on their clay minerals. The LL and PL of some CMWGs reported in the literature are listed in Table 2.

Also included in the table are data from this study related to coal heap samples obtained from the Forjas Santa Barbara mine in Spain.

Table 2: LL, PL, I_p of some weathered aggregates and washery wastes. The letters C, G, L, M, H, S, W stand for clay, gravel, low plasticity (for silt) or lean (for clay), silt, high plasticity, sand, well-graded, respectively, in the unified soil classification system.

Unified soil classification system/*Gs*	**LL**	**PL**	**I_p**	**Location**	**Reference**
GM/2.65	42	25	17	Jerada, Morocco	(Amrani et al. 2020)
GW-GC/(not available)	34.1	22	12.1	Queensland, Australia	(Gallage et al. 2015)
GW-GC/1.85	37.1	26.3	10.8	Enugu, Nigeria	(Okogbue and Ezeajugh 1991)
GW-GM/(2.53-2.75)	21.1	non-plastic	non-plastic	Matallana de Toríno, Spain	(Cadierno et al. 2014)
GM/(2.23-2.76)	19.9	non-plastic	non-plastic	Matallana de Toríno, Spain	(Cadierno et al. 2014)
GM/(2.27-2.76)	22.4	14.9	7.5	Llombera, Spain	(Cadierno et al. 2014)
GW-GM/(2.55-2.74)	17.1	14.5	2.6	Santa Lucía, Spain	(Cadierno et al. 2014)
GM/(not available)	18.8	non-plastic	non-plastic	Cinĕra, Spain	(Cadierno et al. 2014)
GW-GM/(not available)	23.3	19.7	3.6	La Robla, Spain	(Cadierno et al. 2014)
CL/2.72	38	20	18	Indiana, USA	(Jung and Santagata 2014)
SM/2.59	31.5	24.0	7.5	Forjas Santa Barbara mine, Spain	This study
SW/2.13	27.2	17.7	9.5	Wollongong, Australia	(Rujikiatkamjorn et al. 2013)
GM-MH/(not available)	40-73	30-54	10-19	Eskihisar strip coal mine, Turkey	(Ulusay et al. 1995)
ML/1.61	41.7	33	7.7	Site no. WDH-1, USA	(Busch et al. 1975)
ML/1.60	38	35.3	2.7	Site no. WDH-2, USA	(Busch et al. 1975)
ML/1.58	34.3	non-plastic	non-plastic	Site no. BDH-1, USA	(Busch et al. 1975)
MH/1.87	51.1	38	13.1	Site no. BDH-2, USA	(Busch et al. 1975)
CL/2.59	32.3	13.6	18.7	Dinajpur, Bangladesh	(Hossain et al. 2018)
CL/2.63	26.9	17.1	9.8	Basundhara opencast coal mine, India	(Mallick and Mishra 2017)

The data from Table 2 are plotted on Casagrande's plasticity chart overlaid with locations of common clay minerals in Figure 7. Casagrande (1948) suggested this plot as an approximate way to identify the dominant mineral groups present in soils (Holtz and Kovacs 1981). Data from Table 2 are shown to plot primarily on the lower left corner of the chart, indicative of materials that do not hold water well and exhibit non-plastic to moderately plastic behaviours. Mineralogical analysis of the lean clay (CL) from Dinajpur (Bangladesh) was not reported by Hossain et al. (2018) but the California bearing ratio test results showed that it has an expansion ratio of 1.51 thus was unsuitable to be recycled in a road subgrade. X-ray diffraction (XRD) analysis of the sample from the FSB mine in Spain shows that the clay minerals in it comprise of 20% illite and 10% kaolinite, which are non-expandable, and 5% vermiculite which has limited expansion capacity.

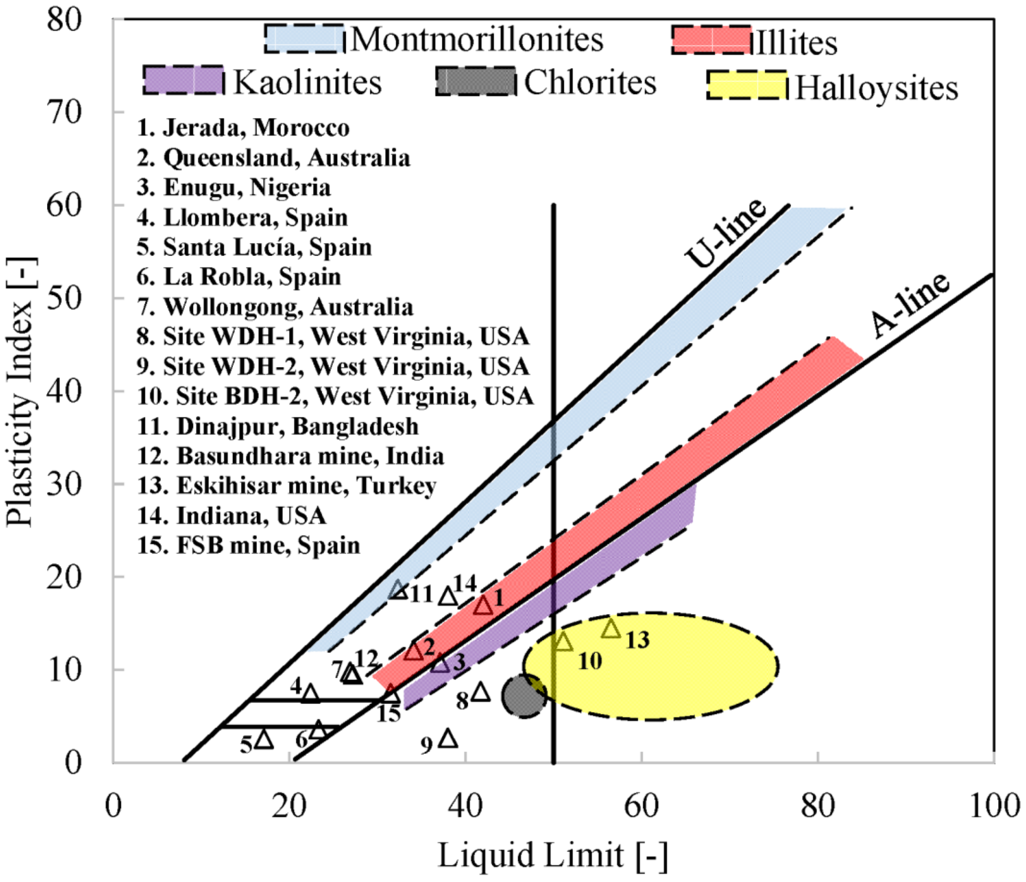

Figure 7: CMWGs' plasticity and clay minerals indicated on Casagrande's chart

4.2.4. Grain size and shape, and grain size distribution (GSD) of CMWGs

The shape of individual granular particles (with particle sizes > 63 μm) is at least as important as the grain size distribution in governing their engineering response (Holtz and Kovacs 1981). Many measures of shape exist, at the most basic level of description, and a useful distinction can be made here between needlelike/flaky particles and bulky particles (Holtz and Kovacs 1981; Rodriguez et al. 2012). The percentage of flaky particles in a given soil tends to increase with decreasing grain size as a consequence of the geological processes of soil formation (Terzaghi and Peck 1948). When being compressed, needlelike/flaky particles compact more than bulky particles (Holtz and Kovacs 1981; Penman 1971); and when being sheared, their different shapes contribute differently to frictional resistance (Holtz and Kovacs 1981; Terzaghi and Peck 1948). In particular, particle shapes strongly affect how materials can be mixed and compacted. As particles become less bulky, both the maximum void ratio $e_{\max}$ and minimum void ratio $e_{\min}$ increase, and $e_{\max}$-$e_{\min}$ also increases (Cho et al. 2006; Cubrinovski and Ishihara 2002; Fraser 1935; Santamarina and Cho 2005). In terms of size and grading, it has been found that when two or more granular soil samples of the same mineralogical content are compacted to the same density under the same effective confining stress, as D_{50} (grain size for 50% finer by weight on a GSD plot) increases, the peak shear strength and volumetric dilatancy decrease; and for soils with the same D_{50}, a less uniform grading (i.e., higher coefficient of uniformity C_u) yields a slightly lower peak shear strength (Harehdasht et al. 2018; Kirkpatrick 1965). However, when sheared to constant volume condition, the shear strength of granular soils depend primarily on the mineral compositions of the particles (Muir Wood 1990; Negussey et al. 1988).

The ways coal-bearing rocks are mechanically broken and mined, and the treatment of left-overs are consequential to properties of CMWGs. This can be reflected on a grain size distribution (GSD) plot. The GSDs of many geomaterials show a self-similar (fractal) distribution (Perfect and Kay 1995). Researchers demonstrated that the breaking up of larger clusters/particles by mechanical actions results in smaller clusters/particles, the resulting GSD exhibits fractal characteristic (Coop et al. 2004; McDowell et al. 1996). Tang et al. (2014) conducted sieving experiments on 30 kg samples of coal gangues and found that their GSDs exhibit fractal characteristics. Yang et al. (2021) conducted drop weight tests of coal samples and found that the GSDs of broken fragments exhibit fractal characteristics. Ding and Liu (2021) immersed a soft slate in water for different durations and found that it disintegrates into particles with GSDs obeying different fractal distributions. Latest studies show that the GSDs of many mine tailings (Qiu and Sego 2001; Vo et al. 2020) and coal tailings (Islam 2021; Salam et al. 2019; Vidler et al. 2020) also exhibit fractal characteristics. Russell (2010) described a GSD exhibiting a single fractal scaling as:

$$\%M_s(L < d_s) = 100\left(\frac{d_s^{3-D_s} - d_{s\,min}^{3-D_s}}{d_{s\,max}^{3-D_s} - d_{s\,min}^{3-D_s}}\right) \tag{4}$$

where D_s, d_s, $d_{s\,max}$, $d_{s\,min}$, M_s denote, respectively, fractal dimension of a GSD, a specific grain size, the maximum grain size, the minimum grain size, and dry mass of particles.

GSDs of some coal aggregates and tailings reported in the literature are replotted in Figure 8 to show how they could be approximated by Eq. (4). Figure 8 shows that a well-graded GSD curve corresponds to a higher D_s and a poorly-graded GSD curve corresponds to a lower D_s. Also plotted in the figure is this study's data obtained from the FSB mine in Spain. The GSD of this sample obeys a single fractal scaling law with $D_s \approx 2.63$.

Eq. (4) may be extended to describe a GSD exhibiting double fractal characteristics as:

$$\%M_s(L < d_s) = \frac{M1}{M1+M2}100\left(\frac{d_s^{3-D_{s1}} - d_{s\,min1}^{3-D_{s1}}}{d_{s\,max1}^{3-D_{s1}} - d_{s\,min1}^{3-D_{s1}}}\right) + \frac{M2}{M1+M2}100\left(\frac{d_s^{3-D_{s2}} - d_{s\,min2}^{3-D_{s2}}}{d_{s\,max2}^{3-D_{s2}} - d_{s\,min2}^{3-D_{s2}}}\right) \tag{5}$$

where $M1$ is the mass percentage of population 1 (fractal dimension D_{s1}), $M2$ is the mass percentage of population 2 (fractal dimension D_{s2}), and $d_{s\,min1} < d_{s\,max1}$, $d_{s\,min2} < d_{s\,max2}$. D_{s1} can be approximated on a GSD plot but D_{s2} would need to be identified by upscaling its mass percentage to 100%. Figure 8 shows how D_{s1} may be approximated from the GSDs of samples exhibiting double fractal characteristics. The samples of Yu et al. (2019) and Qiu and Sego (2001) with GSDs on this figure were collected *in situ* from an Appalachian coalfield in Kentuckys (USA) and from a coal wash plant in the Coal Valley mine in Alberta (Canada), respectively.

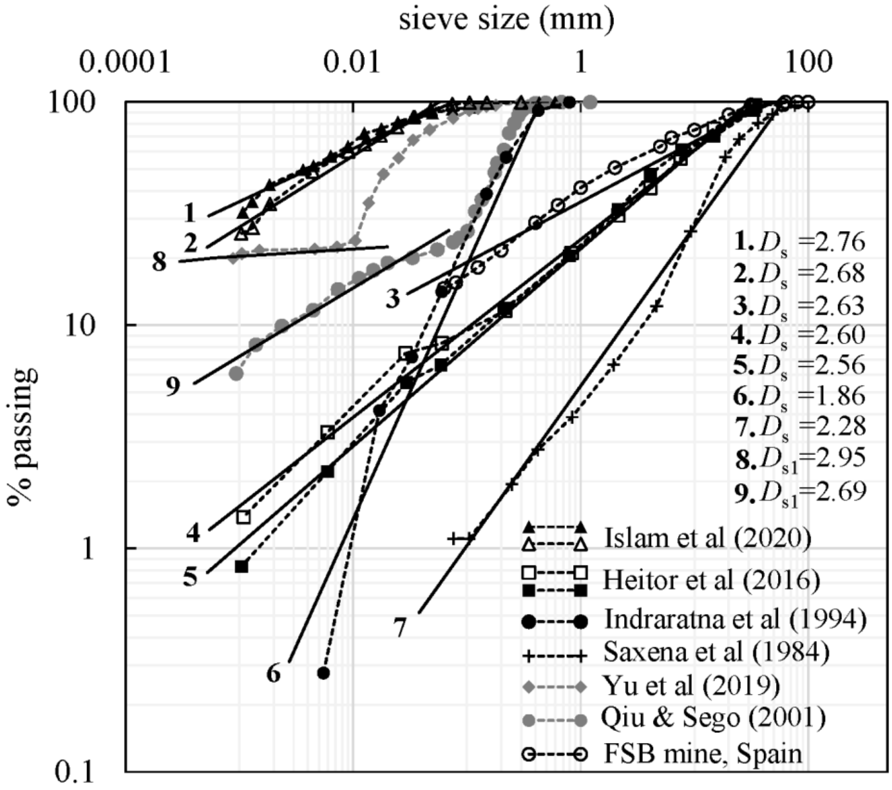

Figure 8: Fractality and heterogeneity in GSDs of CMWGs

Knowing the GSD is useful for estimating the mass and volume of aggregates needed to make CMWG-bearing construction products. This can be made even simpler when the GSD obeys a single or double fractal scaling law. However, there is a high degree of heterogeneity within the CWMGs in spoil heaps. Coal mining wastes weather rapidly when exposed to the elements

(Bishop 1973; Skarżyńska 1995a) but may remain relatively intact when buried deeply within an unburnt spoil heap (Taylor 1975), thus depending on various factors (e.g., how the original wastes were processed and deposited, how long they have been left there, whether they were disturbed during storage) the GSDs of CMWGs reclaimed from storage may eventually be more complex.

4.3. Mineralogical and state-dependant properties of CMWGs

4.3.1. Water retention and hydraulic conductivity of CMWGs

The saturated hydraulic conductivity (*Ks*) of intact coal mining waste aggregates varies depending on the *Ks* of their parenting rocks; e.g. approximately 10^{-4}-10^{-8} cm/s for sandstone, 10^{-7}-10^{-11} cm/s for shale, and smaller for unfractured metamorphic and igneous rocks (Bear 1972; Freeze and Cherry 1979). The very low *Ks* of intact coal mining waste aggregates can be attributed to their small pore sizes, i.e. generally smaller than 50 nm (Mastalerz et al. 2012; Ma et al. 2017; Li et al. 2019). However, when coal mining waste geomaterials are deposited in spoil heaps or impoundment facilities (as CMWGs), or processed into construction products, it also becomes relevant to consider *Ks* of an aggregation of coal mining particles (in addition to *Ks* of intact aggregates themselves). The *Ks* of CMWGs is dependent on their grain and pore size distributions, and compactness (Holubec 1976; Leventhal and de Ambrosis 1985; Skarżyńska 1995a; Ulusay et al. 1995). The value of *Ks* for CMWGs vary widely because these geomaterials are susceptible to rapid weathering (Bishop 1973; Cobb 1977; Holubec 1976; Saxena et al. 1984; Skarżyńska 1995a; Taylor and Spears 1973) and fabric inhomegenity (Cobb 1977; Kirby 1980; Saxena et al. 1984). An increase in the degree of weathering is associated with an increase in the portion of finer particles and pores and a decrease in *Ks*. Freshly wrought coal waste aggregates deposited loosely in spoil heaps can be very permeable with *Ks*=10^{-1}-10^{-2} cm/s (Skarżyńska 1995a), but with weathering and different levels of compaction, *Ks* can be anywhere between 10^{-1} to 10^{-8} cm/s for coal waste aggregates (Holubec 1976). Entrainment of fines at the interface between a tailing lagoon and its embankment may reduce Ks down to 10^{-12} cm/s, i.e., effectively impermeable.

It was found that Ks of CMWGs measured *in situ* in the UK are about two orders of magnitude higher than those measured in the laboratory (Cobb 1977; Kirby 1980). Hence the UK-based studies recommend a greater reliance on *in situ* measurements (Murray and Symons 1974; National Coal Board 1972). Saxena et al. (1984) found the average Ks measured *in situ* (on a site in the USA) to be somewhat higher than in the laboratory and attributed this difference to fabric. They found that with decreasing lift thickness, the field permeability decreases. Rujikiatkamjorn et al. (2013) found that compacting the coal wash at wet and dry sides of the optimum moisture content results in samples with different fabrics, and Ks versus void ratio relations. For coal tailing deposits, distinct layers of different physical compositions are often noticeable on visual examination.

With the exception of those wastes buried below ground water level to manage the AMD problem, CMWGs *in situ* are generally unsaturated. Recent studies (Alonso and Cardoso 2010; Oldecop and Rodari 2017; William 2012) showed a wide scope of applications of geomechanics of unsaturated media in coal mining and post-mining operations. Geomechanics of unsaturated media can be applied to characterise behaviors of CMWGs and porous construction products containing them. In particular, water retention curve and hydraulic conductivity function could be obtained to quantify how fluids and gases move through the pore spaces. To show this simply, the 1D steady state version of Darcy's law (Buckingham 1907; Griffiths and Lu 2005; Richard and Fireman 1941) can be expressed as:

$$q = -K\left(\frac{d\psi}{dz} + 1\right) \tag{6}$$

where q is the flow rate (cm/s) and $d\psi/dz$ is the pressure gradient driving flow in the z direction. Assuming $\psi=u_a-u_w\equiv s$ (kPa) where u_a, u_w, $s\equiv$ pore air pressure, pore water pressure and matric suction, respectively, then i) the water retention curve is $s=f(s_e, Sr, e, \text{etc.})$ where $s_e\equiv$air entry/expulsion suction, $Sr\equiv$ degree of saturation, $e\equiv$ void ratio, and ii) the unsaturated hydraulic conductivity function is $K=f(Ks, Sr, e, \text{etc.})$. There are many empirical models of water retention curve and hydraulic conductivity function in the literature, adopting them often requires calibration against relevant experimental data.

Water movements induce volumetric changes in intact coal mining aggregates, a behaviour found to be strongly dependant on coal rank and pore characteristics (Suuberg et al. 1993; Stanmore et al. 1997; McCutcheon et al. 2001; Ma et al. 2016). However, there are limited experimental studies on water retention characteristics of aggregations of coal waste particles (as CMWGs). Sharma et al. (1993) mixed lumps of coal and soil together in different ratios to create coal spoil samples then tested them in a pressure plate device. They reported different water retention behaviours between samples containing commercial lignite (with high water repellency) and samples containing degraded lignite (with low water repellency). Qiu and Sego (2001) studied the water retention characteristics of a coal tailing using the pressure plate test. The air entry value was reported to be 18 kPa which is somewhat low considering that the material was classified as a low plasticity clay. Residual volumetric water content was 18% which is rather high. Fityus and Li (2006) conducted filter paper tests of processed Australian coals and reported a significant difference between the soil water retention curve of the coals and of a typical soil due to the coals' strong hydrophobic nature. They concluded that on drying from a saturated state, processed coal would have negligible suction until Sr < 0.5-0.6, hence much of the moisture in stockpiled coal would not contribute to forming films and adhesion to suppress the release of dust. Vidler et al. (2020) obtained the soil water retention curves of a coal tailing, the mineral fraction and the coal fraction using a tensiometer and a dewpoint potentiometer. They found that the presence of coal in the tailing has a significant impact on the water retention behaviour of the coal tailing. The coal fraction desaturates at low suction on drying from a fully wetted state, its inclusion in tailing might cause localized hydrophobicity, and overall lower the air entry value of the geomaterial. Liu et al. (2021) investigated in experiments the effect of drying-wetting cycles on the hydromechanical behaviour of a compacted coal gangue. They found the pore-size distribution curve of a coal gangue to exhibit a bimodal feature. Both the inter-aggregate pores and intra-aggregate pore volumes were found to be affected by hydraulic loading.

4.3.2. Shear strength of CMWGs

As it is customary and widely adopted in practical geomechanics field, Mohr-Coulomb parameters are adopted here to discuss the shear strength of CMWGs. The shear strength parameter τ can be defined in terms of total and effective stresses as:

$$\tau = c + \sigma \tan \varphi \qquad (7)$$

$$\tau = c' + \sigma' \tan \varphi' \qquad (8)$$

where c, σ, φ, c', σ', φ' denote total cohesion, total stress, total friction angle, effective cohesion, effective stress, effective friction angle, respectively. The effective stress (σ') for saturated and unsaturated CMWGs can be defined respectively as (Bishop 1959; Terzaghi 1943):

$$\sigma' = \sigma - u_\mathrm{w} \qquad (8)$$

$$\sigma' = (\sigma - u_\mathrm{a}) + \chi(u_\mathrm{a} - u_\mathrm{w}) \qquad (9)$$

where $\chi \equiv$ effective stress parameter (χ=1 for fully saturated conditions and χ=0 for fully dry conditions). For unsaturated geomaterial, u_a-u_w=s. The $\chi(u_a$-$u_w)$ component in Eq. (9) contributes to the effective stress and shear strength of unsaturated geomaterials. Bishop (1960) suggested that χ depends on many factors including Sr, s, the drying-wetting cycle and the stress history of the geomaterial. Characterising the dependency of $\chi(u_a\text{-}u_w)$ to different states is the key to estimate the shear strength of unsaturated geomaterials.

Different types of shearing tests and interpretations of test data contributed to a large variation of c', φ' for CMWGs reported in the literature (Holubec 1976). This section will focus on triaxial shearing tests.

The shear strength of saturated CMWGs has been extensively characterized in triaxial shearing tests. Typical c', φ' of CMWGs obtained from consolidated undrained (CU), consolidated drained (CD), multistage consolidated undrained (m-CU), multistage consolidated drained (m-CD) tests reported in the literature are shown in Table 3. Also included, is this study's data of a coal heap sample obtained from the FSB mine (Spain). The shear strength of a CMWG varies considerably depending on its sampling location (Bishop et al. 1969; Kirby 1980). The c', φ' in Table 3 were the peak shear strength parameters for the level of shear strain, compactedness and effective confining stress considered in those studies. The c', φ' in Table 3 do not differ significantly between coal tailing and coal waste aggregates. The shear strength of coal tailing in Appalachian region, USA was found to be relatively high given its grain size distribution (Holubec 1976). Thompson et al. (1973) and Taylor (1974b) attributed the relatively high shear strength of coal tailing to the presence of the coal mineral in it. Kirby (1980) found that both the coal content and shear strength of coal tailings in the UK were in fact higher than those of the coal waste aggregates.

Table 3. (c', φ') obtained from consolidated undrained (CU), consolidated drained (CD), multistage consolidated undrained (m-CU), multistage consolidated drained (m-CU) tests on saturated samples

Type	**Unified Soil Classification System/*Gs***	***c'* (kPa)**	***φ'* (°)**	**Test**	**Location**	**Reference**
Impounded waste	(not available)/1.75-2.22	0	22-39	CD	Aberfan, UK	(Thompson and Rodin 1972)

Impounded waste	(not available)/1.61-2.01	0-20	25-42.7	CU	Peckfield, UK	(Kirby 1980)
Impounded waste	(not available)/1.42-2.32	0-7	30.5-35	CU	East Hetton Colliery, UK	(Kirby 1980)
Impounded waste	(not available)/1.42-2.32	0-27	31.8-40.5	CU	Maltby, UK	(Kirby 1980)
Impounded waste	CL/1.94	10	32	CU	Coal Valley Mine, Canada	(Qiu and Sego 2001)
Impounded waste	(not available)/1.34-1.66	0-14.34	32-40	CU	Appalachian Region, USA	(Holubec 1976)
Impounded waste	SM/2.10	13.8	29	m-CU	Appalachian Region, USA	(Salam et al. 2019)
Impounded waste	ML/2.10	25.5	26	m-CU	Appalachian Region, USA	(Salam et al. 2019)
Impounded waste	ML/2.10	24.8	30	m-CU	Appalachian Region, USA	(Salam et al. 2019)
Impounded waste	SM/2.10	20.7	31	m-CU	Appalachian Region, USA	(Salam et al. 2019)
Impounded waste	SM/2.20	16.5	36	m-CD	Appalachian Region, USA	(Salam et al. 2019)
Impounded waste	ML/2.20	13.1	38	m-CD	Appalachian Region, USA	(Salam et al. 2019)
Coal waste aggregate	(not available)/1.80-2.70	0	23-42	CD	Aberfan, UK	(Thompson and Rodin 1972)
Coal waste aggregate	(not available)/1.75-2.50	0-24	28-41	CU	Appalachian Region, USA	(Holubec 1976)
Coal waste aggregate	GM-MH/(not available)	0-10	23-35	CD	Eskihisar coal mine, Turkey	(Ulusay et al. 1995)
Coal waste aggregate	GW-GC/(not available)	1-40	33.2-38.6	m-CU	Queensland, Australia	(Gallage et al. 2015)
Coal waste aggregate	SC/2.23	0	35	CD	NSW, Australia	(Heitor et al. 2016)
Coal waste aggregate	(not available)/(not available)	5-25	33-36	CD	QLD, NSW & VIC, Australia	(Simmons 2020)
Coal waste aggregate	SM/2.59	0	29.1	CU	Forjas Santa Barbara mine, Spain	This study

There are few laboratory experiments investigating the effective shear strength parameters (c', φ') of unsaturated CMWGs. Most experiments conducted on unsaturated coal mining wastes obtained the total strength parameters (c, φ). Eqs. (7)-(9) show that the separate impact of u_a, u_w and χ on shear strength cannot be distinguished using the total strength parameters c, φ.

Saxena et al. (1984) reported that the unconfined compression strength of CMWGs was more dependent on the water content at the time of testing rather than on dry density. Okogbue and Ezeajugh (1991) investigated an unsaturated Nigerian coal waste in experiments and obtained φ=13.4°-15° and c=55-57 kPa in UU (unconsolidated undrained) test, and c=38.6-39.2 kPa in UC (unconfined compression) test. The relatively high c and low φ were attributed to the type of tests (i.e., UU, UC). If the test data for Forjas Santa Barbara mine (Spain) (Table 3) were to be interpreted in total stress, c=111 kPa>c'=0 kPa, φ=27.7°<φ'=29.1°. Indraratna et al. (1994) showed that φ of compacted coal tailings obtained from unsaturated CU tests was maximum at optimum moisture content although the influence of void ratio was not accounted for separately in their results.

Particle breakage is another factor that affects the shear strength of weak coal-bearing rocks and aggregates and their potential for reuse. It has been established that many types of aggregates when used as rockfills (including coal-bearing aggregates) are prone to particle breakage once the effective confining stress reached a critical value (Marachi et al. 1972; Marsal 1967, 1973). Indraratna et al. (1998) tested latite basalt aggregates (flakiness index FI=25%, D_{50}=30-40 mm, C_u=1.5-1.6) in a large triaxial test to characterise their shear behaviour as railway ballast. They reported a departure in shear and deformation behaviours of the basalt between low effective confining stress (<100 kPa) and higher effective confining stress. Breakage was found to be influenced by the shape, size, grading of particles and the compactedness of test samples. At higher levels of effective confining stress, localized breakage occurred at contact points between particles. The contact stress can be much higher than the applied deviator stress. Broken particles fill up the void spaces and reduce the hydraulic conductivity of the porous medium (Ma et al. 2017). In practice, this mechanism of particle degradation is commonly observed to lead to clogging and undrained failure of railway ballast (Chrismer and Read 1994). Heitor et al. (2016) tested a coal wash from Wollongong

(Australia) in drained and undrained triaxial compression tests in effective confining stresses of up to 600 kPa, and isotropic compression tests in effective confining stresses of up to 1,400 kPa. It was found that the compaction of the coal wash aggregates at their natural moisture content into triaxial samples induced significant particle breakage. The aggregates compacted under 170, 341 and 681 kJ/m^3 (using standard Proctor compaction device) each attained unique effective strength parameters *(c', φ')* at the critical state. Consolidation and shearing of the coal wash also induced significant particle breakage when a critical effective confining stress (in that case, 127 kPa) was exceeded.

5. Properties relevant to the recovery of metals from CMWGs

Unprocessed coal contains both organic and inorganic matter, which can host a wide range of major and trace metals. Geological processes operating during peat accumulation and diagenesis, including volcanic ash, sediment deposition, or infiltration by subsurface fluids, can contribute metals that are not originally present in the peat itself. Consequently, several rare (e.g., Ga, Ge, U, Nb, Zr), precious (e.g., Au, Ag) and base (e.g., Al, Fe, Mg) metals can be enriched within coal-bearing strata, and these have been known to reach concentrations that are comparable with some conventional ore deposits. Such metals can be present in a wide range of physical and chemical forms, which include, being hosted in fine grained minerals that are directly encased within the coal-bearing matrix; being sorbed onto the surface of organic matter; and being dissolved in pore waters (Dai et al. 2020). Consequently, CMWGs derived from such ‘metalliferous’ coal seams have the potential to be a source for metals whose primary extraction process is expensive, or which are especially limited in supply.

To date, commercial metal extraction from coal has been limited to its combustion products; the metals being obtained as byproducts of thermal power generation. Such extraction has been undertaken at industrial scale for U and Ge, at a more modest scale for Au and Ag, and at pilot scale for Al, Ga, Si, Mg and some of the rare earth elements (REEs) (Dai and Finkelman 2018). U coal mines were undertaken by the USA and the USSR during the rapid initial development of the nuclear industry after the Second World War. Coal-hosted U is predominantly bound to organic matter, and although coal is no longer the primary U source, interest has recently revived amid concerns about the depletion of conventional deposits (Dai and Finkelman 2018). Ge has been sourced from coal since the 1950s, and even today, Russian and Chinese coal mines account for more than half of global production (Seredin et al. 2013). Like U, Ge is predominantly hosted by organic constituents of the coal (Wei and Rimmer 2017), and is liberated upon combustion and recovered in purpose-built ash collectors. At a much smaller scale, Au and Ag have been mined from North American coals during the 19th and 20th centuries (Seredin and Finkelman 2008), though neither metal is appreciably sourced in this manner today. Pilot plants, built as technology demonstrators for the recovery of Al and Ga and the REEs have been under development in Mongolia and the USA (Dai and Finkelman 2018, Honaker et al. 2019).

Commercial metal extraction from CMWGs (rather than combustion products) has no historical precedent of which the authors are aware. Currently, the most intensively studied metals in this regard are the REEs, which are frequently more enriched in coals than they are in crustal rocks on average (Zhang and Honaker 2019). This enrichment has been attributed to the chelating ability of humic acids that are present in nascent seams during the coalification process (Zhang and Honaker 2019), and to the various geological processes of sediment redistribution, volcanic ash deposition, and groundwater and hydrothermal infiltration (Hower et al 2016). Average cumulative REE concentrations (i.e., for all of the lanthanides plus Sc and Y) in coal have been reported as 69 and 72ppm in lignite and bituminous coals respectively (Ketris and Yudovich 2009), although considerable variability occurs among the world’s coal seams. In extreme cases, such as the Fire Clay seam in East Kentucky (USA), these cumulative concentrations can be as high as 4540ppm (Zhang and Honaker 2019). Notably, coal and coal derivatives contain a greater proportion of the more commercially valuables REEs then do primary ores; the heavy REEs (HREEs: Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu) and 'critical' REE (CREEs: Nd, Eu, Tb, Dy, Y) (Seredin and Dai 2012).

In addition to the abundance of REEs in CMWGs, the manner in which they are incorporated into these waste materials is critically important in determining their viability as a commercial resource. For example, REEs adsorbed onto mineral surfaces are more readily extracted by leaching than those hosted inside mineral lattices. Since minerals exhibit a wide variability in solubility certain minerals require more expensive and/or resource intensive leaching agents and associated leaching conditions (e.g., high temperature and/or pressure) to extract the REEs hosted in their lattices. Indeed, the 'mode of occurrence' of REEs in CMWGs remains a vigorous area of research (e.g., Finkelman et al. 2018). Recent studies have typically applied optical and scanning electron microscopy (SEM) to identify the REE-bearing particles directly, and such research is often coupled with mineral phase analysis (e.g., QEMSCAN (quantitative evaluation of materials by scanning electron microscopy), XRD, Raman Spectroscopy) and sequential chemical extraction experiments. The latter technique is employed to quantify the solubility and physical deportment of metals within the CMWG for leaching. It typically consists of the application of a series successively more oxidizing leaching agents, so that more strongly-bound metals are progressively removed. The metal concentrations of the leachates obtained can be used to calculate the proportion of each metal that is bound to the various components (or 'fractions') of the sample. Sequential extraction data (e.g., Zhang and Honaker 2019, Zhang and Honaker 2020, Finkelman et al. 2018) have indicated that in general, REEs are dominantly bound to the insoluble oxide or silicate fractions of CMWGs, indicating that they are generally hosted by minerals that may only be dissolved using strong mineral acids. SEM analyses have identified these minerals as monazite, xenotime, zircon, florencite, allanite, apatite (calcium phosphate), alunite supergroup minerals, hydrated phosphates, and less commonly, oxides, carbonates or fluorocarbonates (Dai and Finkelman 2018).

It must be emphasized that there are exceptions to the above general conclusions, which arise from the diversity in both coal rank and REE geochemical behaviour. For example, Finkelman et al. (2018) reported that a higher proportion of HREEs are organically bound than LREEs, and a study of lower-rank lignite coals by Laudal et al. (2018) indicated all of the REEs were dominantly bound organically. However, for the majority of CMWGs investigated to date, the REEs have been typically recorded as mineral-hosted and therefore difficult to economically separate from the gangue material. As a result, extraction of REEs from CMWGs presents a formidable technical challenge, potentially requiring pre-treatment of the waste material by heating or physical beneficiation (mechanical separation on the basis of density or hydrophobicity) before the target metals can be leached. For example, at a pilot plant developed to extract REEs from North American coals, it was reported by Honaker et al. (2019) that heating to 600°C for 2 hours raised the fraction of REEs that can be leached with H2SO4 from 25% to 85%. The commercial viability of CMWGs as a source of REEs will depend on the success of future treatment processes, as well as the development of appropriate leaching agents.

6. Conclusions

There is currently a drive to develop innovative concepts for managing, recycling and upcycling waste geomaterials generated by coal mining activities in Europe and throughout the world. CMWGs present us with many challenging problems such as spontaneous combustion and leaching of acidic water to the surrounding environment, slope instability of spoil heaps and flow liquefaction of impoundment facilities. Storing CMWG deposits consume economic resources yet there is great demand of raw geomaterials in the construction industry. For example, 2.7 billion tonnes of natural aggregates are produced in Europe for construction purposes every year, although there is an imperative to conserve natural resources.

This paper reviewed the properties of CMWGs relevant to assess their suitability as raw geomaterials in construction industry. Many features of this review set it apart from other reviews. This review integrates previous understandings with state-of-the-art literature from geochemical, geotechnical and structural engineering to provide a multifaceted view of CMWGs' properties. This is essential because historically coal mining wastes have been regarded as problematic geomaterials thus there is resistance to their utilisations in the communities. Secondly, the review does not only focus on properties of the geoparticles themselves but also on properties of their surrounding pore and solid networks. This is essential because the geoparticles have to be reclaimed from CMWGs' storage facilities, further processed into components of construction products. In those conditions the geoparticles are aggregated where interactions with the surrounding pores and particles may govern their macrobehaviours. Also, due to the complexity of CMWGs' properties, this review attempted to distill from the literature some practical yet simple steps that yield useful information for the initial and more substantive screening of CMWGs in specific construction applications.

- With regards to geochemical aspects of CMWGs, it was emphasized previously in the literature (e.g., Younger 2004) that coal mining wastes are associated with some major problems such as air pollution, fire hazards, ground deformation, water pollution, and water re-source depletion. Assessing the suitability of CMWGs for upcycling as a secondary raw material for higher level civil engineering applications requires a robust understanding of the geochemical processes it is likely to undergo. The suitability of CMWGs for a particular construction application can vary from one mine to another due to their diverse mineral contents. It was found in this review that CMWGs are good SCM candidates because they often contain large fractions of clay minerals that can be conditioned (and blended with other materials when necessary) to acquire pozzolanic properties, as has been demonstrated in many recent studies (e.g., Bich et al. 2009; Frías et al. 2012; Vigil de la Villa et al. 2014; García Giménez et al. 2016; Caneda-Martínez et al. 2019; Rodríguez et al. 2021). It was also concluded in this review that more research is needed to investigate the propensity for CMWGs to promote ASR (since this condition can take years to realise) and ACR (since the extent to which this reaction impacts durability is not sufficiently delineated from other processes). Furthermore, it was found that the recovery of metals from CMWGs is challenging and worthy of further investigation. The commercial viability of metal recovery from these wastes depends critically on their relative abundance and the manner in which they are incorporated into the host particles. Selective recovery of metals from CMWG-bearing construction products appears to be a much more feasible option.

- Previous reviews (Holubec 1976; Skarżyńska 1995a; Masoudian et al. 2019) emphasized that CMWGs are chemically and physically highly-heterogeneous, and CMWGs have been utilised successfully in many geotechnical and specialised structural applications (Hammond 1988; Skarżyńska 1995b; Liu and Liu 2010). This review focuses on the interactions between mineralogy, geotechnical indices, and state-dependant properties of CMWGs. It was found that the mineral content of a CMWG can influence both the durability and the strength of its potential construction applications. Simple techniques are provided to aid the initial screening of CMWGs for their suitability e.g., the heterogeneity of a CMWG may be predicted via its GSD, the carbon content of a CMWG can be approximated by its Gs, the presence of major clay minerals in a CMWG may be indicated by how LL, I_p plots on a Casagrande’s chart. When a more substantive screening of CMWGs for suitability is needed, it was found that quantifying the amount of expandable minerals and durability performance using techniques such as x-ray diffraction and slake durability test is important. Moreover, it was found that the highly-heterogenous state-dependent properties of CMWGs (e.g., water retention, hydraulic conductivity, shear strength) are impacted by complex factors such as coal content, particle size and shape, pore scale spanning 6-9 orders of magnitude, and the presence of pore air and pore water in the interstitial void space. In this respect, recent studies (Alonso and Cardoso 2010; Oldecop and Rodari 2017; William 2012) showed that there is a wide scope of applications of geomechanics of unsaturated media in characterising the behaviors of CMWGs and porous construction products containing them.

- There are still scattered experiences with reference to the application of CMWGs as constituents of concrete and cement based mixtures either as supplementary cementitious material replacing Ordinary Portland Cement or as recycled aggregates in substitution of natural ones. Surveyed studies have highlighted on the one hand the importance of proper mineralogical and mechanical characterization of CMWGs for their suitability to the purpose above. On the other hand, the need and possibility have been demonstrated of finding, through appropriate tests, the optimal replacement percentages in order to obtain a concrete mix featuring the level of mechanical and durability performance required for the intended engineering application. Variations, generally reductions, due to the incorporation of CMWGs can be kept within limits which still make the obtained concretes suitable for the intended purposes through appropriate selection and grading of the same CMWGs and suitable mix-design approaches, highlighting the importance and need of a unified framework for promoting their valorization into cement based construction materials and products.

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Declaration of Interest Statement

Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: