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OPPORTUNITIES OF WET-HANDLED COAL BOTTOM ASH USE IN BINDING MATERIALS: A REVIEW

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Nowadays conventional binding material for the construction sector is Portland cement. Portland cement consists mainly of high-energy intensive with a significant carbon footprint Portland cement clinker. Reduction of clinker content in binding materials becomes the utmost priority for scientists in the field, it is reflected in manufacturers' Sustainability Road Maps. This fact triggers searches and actions in different directions such as improving grinding technologies, chemical additives and admixtures development, and extension of the cementitious portfolio itself to increase the availability of raw materials. More and more often in construction technologies materials that relatively recently did not represent a value as cementitious due to the availability of more easy options, are being used. This article considers opportunities and aspects of wet-handled coal bottom ash use from thermal power stations.

Key words: supplementary cementitious material; wet-handled coal bottom ash; fly ash; portland cement; CO_2-emission; low-carbon binder.

Introduction

The construction industry is one of the most energy-intensive parts of human activity. It accounts for more than one-third of all final energy and half of total electricity consumption worldwide (Attia et al., 2022), this causes high greenhouse gases and other pollutants emissions making a significant contribution to climate change that needs global solutions in all aspects of construction and operations of buildings as well as other infrastructure. Regardless of different investigations in the area of binding materials, conventional Portland cement remains the predominant material and it is anticipated that it will do so in the next number of decades. Portland cement clinker manufacturing generates from 830 to 1200 kg CO_2 per ton of clinker when using fossil fuels and fossil raw materials depending on the process applied. In construction projects, the conventional binding material is Portland cement with clinker content from 60 to 95 %, which corresponds to CEM I, CEM II A, and CEM II B types of Portland cement. In Ukraine State Technical Standards DSTU EN 13282-1:2021 and DSTU EN 13282-2:2021 intend to use multicomponent hydraulic road binders with low clinker content.

Traditional cementitious material in Portland cement is ground granulated blast furnace slag (GGBFS), which is a by-product of steel production. Because of the growing demand for GGBFS and the expected stoppage of some steel-making plants due to their CO_2-intensive technology, GGBFS volume will be far not sufficient to satisfy the construction industry in cementitious materials, thus extending the horizon of other cementitious such as pozzolans is key. One of the under investigated materials in this term is wet-handled coal bottom ash from thermal power stations. Considering its high availability in Ukraine it is important to explore opportunities to use it as a cementitious material.

Prerequisites for the use of cementitious materials to cement

Nowadays conventional binding material for the construction sector is Portland cement. There is a forecast claiming that cement demand will grow from 12 % to 23 % by 2050 due to population increase and future urbanization (Khan et al., 2022). In Ukraine, an increase in cement consumption is expected due to postwar reconstruction. Such challenges are connected to anthropogenic CO_2 emissions and global warming. In its Green Deal, the European Union sets out its ambition for Europe to be the first climate neutral continent by 2050. This requires robust actions in all sectors of economics, in particular in the construction sector, which is a source of approximately 5 % of all emissions in the world. Therefore European Cement Industry Association CEMBUREAU published the 2020, Road Map to 2050 (2050 Carbon Neutrality Roadmap).

To assess the carbon footprint of the cement sector it is necessary to analyse the chain of manufacturing process from raw materials extraction to concrete use and construction activities (5C approach – Clinker-Cement-Concrete-Construction-Carbonation). At each stage technical solutions are elaborated to reduce greenhouse emissions. The main component of Portland cement is clinker, which generates a significant amount of carbon dioxide due to the burning of fossil fuels and the decomposition of limestone. Therefore, technical measures are established to ensure zero greenhouse gas footprint from cement making process in both direct (from calcination and thermal processes at clinker manufacturing) and indirect emissions (use of electricity at cement production).

In the case of direct emissions primary and secondary measures are differentiated. In the scope of primary measures the following techniques are considered (2050 Carbon Neutrality Roadmap):

(1) use dry process clinker manufacturing as opposed to semi-dry and wet processes;

(2) replacement of fossil raw materials such as limestone with non-carbonate-bound calcium raw materials (air-cooled slag, carbide lime, calcareous fly ash, waste from the structure destruction) (Sanytsky, Sobol, Shcturmay & Khymko, 2011; Scrivener, John & Gartner, 2018);

(3) replacement of fossil fuels with CO_2 neutral biomass from pure biomass fuels (timber and agriculture wastes) and recovered fuels containing biomass (solid recovered fuels originated from municipal, commercial, and industrial wastes);

(4) reduction of clinker content in cement itself.

In the scope of secondary measures Carbon Capture Storage and Utilization are under consideration. Primary measures techniques are to be utilized in the first approach, this enables a reduction to about 50 % of all CO_2 emissions and gradually reduces energy consumption by cement making industry. Being highly capital and operationally expensive secondary measures techniques are to be considered when possible potential of primary measures is depleted for the specific production unit (2050 Carbon Neutrality Roadmap).

Key measure to reduce CO_2 emission at cement manufacturing process is production and wide application of cements intending use of different cementitious and securing properties of product at the same time. In 2017 average clinker content in cement was 77 % (Sroda, 2020), it is expected to reduce it to 74 % and to 65 % by 2050 (2050 Carbon Neutrality Roadmap).

Nowadays most cement used for building and infrastructure contains above 60 % clinker is chiefly cement of types I and II. In Ukraine, clinker content in cement makes up approximately 75–80 %. In Austria with similar climate conditions currently average clinker content in cement is about 70 %. It is anticipated, that by applying different materials, grinding technologies, and chemicals it is possible to reach nearly average of 52 % of clinker in cement (Responsible and visionary: CO_2-Roadmap).

In this respect, CEM II/C (so-called C-cements) is developed with clinker content from 50 % to 65 % within EN 197-1 standard (EN 197-5:2021), as well as a new group of composite cements CEM VI. A typical example of a binder with a high proportion of mineral components is hydraulic road binders, which are produced according to EN 13282-1:2013 and EN 13282-2:2015, corresponding standards have also been adopted in Ukraine DSTU EN 13282-1:2021 and DSTU EN 13282-2:2021. The minimum content of clinker according to these standards is 10 %. For hydraulic road binder normal hardening of strength class N2, the recommended clinker content is 58.5–71.5 % (Stevulova, Strigac, Junak, Terpakova & Holub, 2021). Research of cement based on multi-cementitious compositions proved the required properties of finish products and possibility to use them in concrete (Kuterasińska & Król, 2016;

Sanytsky, Kropyvnytska, Fic & Ivashchyshyn, 2020; Batog, Bakalarz, Synowiec & Dziuk, 2022). On the other hand, the reduction of the clinker factor requires the availability of raw materials volume. According to European norms (EN 197-1:2000 and EN 197-5:2021) and Ukrainian norms (DSTU B EN 197-1:2015) it is possible to use various cementitious materials. Most often these materials are granulated ground blast furnace slag (S), SCM (silica fume, fly ash and pozzolan – V) and limestone (L, LL). Traditional cementitious material in Portland cement is ground granulated blast furnace slag which is a by-product of steel production. Wide use of GGBFS is driven by its easy to use advantages and good impact on cement physical and mechanical parameters such as low water to cement demand and strength development (Snellings, Suraneni & Skibsted, 2023). Because of the growing demand for GGBFS and the expected stoppage of some steel making plants due to their CO_2-intensive technology, GGBFS volume will be far not sufficient to satisfy the construction industry in cementitious materials therefore deployment of other cementitious sources is vitally important. Family of pozzolanic materials are at the forefront of it.

Pozzolans are a group of siliceous and aluminate materials (generally low-Ca aluminosilicates) that in the presence of water, react chemically with calcium hydroxide) to form cementitious hydrates (Snellings, Suraneni & Skibsted, 2023). There are natural (volcanic ash, zeolite, spongolite) and artificial pozzolanic materials (metakaolin, fly ash). Due to the implementation of Green Deal and increased use of renewable energy availability of pulverized fly ash (V and W types) will become limited in long-term prospective.

Such materials as silica (D), and burnt shale (T) have limited availability, process of receiving metakaolin requires a high energy input of 0.35 GJ/t clay (Juenger, Snellings & Bernal, 2019). The use of fossil materials such as limestone (L, LL) or natural pozzolan (P) in cement is less environmentally friendly in comparison to by-products or wastes of industries. Therefore to secure sustainability principles, circular economy and considering the life circle of materials it is important to consider the use of alternative materials such as by-products or waste materials with pozzolanic or hydraulic properties as replacement of clinker in cements.

The article (Tkaczewska, 2019) describes the use of Cement By-Pass Dust as an additive to Portland cement. However, due to the increased content of alkalines and chlorine, its amount in cement does not exceed 5 %. Authors proved the efficiency of using ash rice husk materials (Amin, Hissan, Shahzada, Khan & Bibi, 2019), ash from sawdust (Odubela & Oluwatob, 2022; Teixeiraa, Camõesa & Brancob, 2019) as replacement of Portland cement in concrete, however, such additives are local materials. Promising is use of ash from paper manufacturing rejects containing β-$2CaO{\cdot}SiO_2$ calcium aluminates $12CaO{\cdot}7Al_2O_3$, helenite $2CaO{\cdot}Al_2O_3{\cdot}SiO_2$ as cementitious material (Sobol et al., 2020; Hunyak, Hidei, Sobol & Petrovska, 2023), at the same time such waste requires additional milling and use chemical admixtures to regulate setting time. Authors showed that using paper mill sludge as a replacement for cement is possible at a limited level of 10 %, at this minor strength loss, is detected (Nazar, Abas & Othuman Mydin, 2014). These wastes appear in relatively small volumes, therefore use of wet-handled coal bottom ash from thermal power stations could be considered as one of the most potential materials as cementitious material.

Wet-handled coal bottom ash availability in Ukraine

In 2019 in Ukraine 15 thermal power plants were in operation. Based on 2019 about 360 million tonnes of wet-handled coal bottom ash was stored in dumps and lagoons, it was expected that the volume of stored wet-handled coal bottom ash would be approximately 415 million tonnes by 2035 (Yevropeiska biznes asotsiatsiia, 2021). The average annum volume of generated wet-handled bottom ash is estimated at level 6–7 million tonnes while consumption is 0.5–0.7 million tonnes, 86 % of consumption accounts for cement producers. As the overall significant potential is in place for utilization of wet-handled coal bottom ash in different fields such as cementitious material in cement manufacturing and ready mix channels, alumina rich raw material for Portland cement clinker, filler in ready mix channel and roads construction, energy saving additive in ceramic bricks manufacturing. The level of cement production in Ukraine was at the level of approximately 11 million tonnes cement annum (2021), thus available wet-handled coal bottom ash makes up significant for development opportunities.

Basic aspects of using wet-handled coal bottom ash as cementitious material to reduce greenhouse gas footprint among other measures

For sustainability reasons, special attention is drawn to material that is waste of other industries namely coal fly ashes. Global coal-fired power generation generates more than 750 million tonnes of coal ash annually, but the level of use is less than 50 % of global production (Izquierdo & Querol, 2012). There are two types of coal fly ashes: dry-handled coal ashes (pulverized fly ashes – PFA) and wet-handled coal ashes. PFA has found wide application as cementitious at cement making process thanks to its ready to use properties at cement grinding (low or no moisture, high fineness) while wet-handled coal ash (herein after wet bottom ash) is still not well investigated and extensively used, mainly to its high moisture (usually above 20 %) and coarse particles which would require additional technological preparation before use as cementitious. Particles of wet-handled coal bottom ash are more porous, coarse (fineness modulus is 1.5–3.8) and of irregular shape in comparison to particles of pulverized fly ash, but chemical content is not very different (Cheriaf, Cavalcante & Pera, 1999).

According to different standards of fly ash use in China, Japan, the USA and the EU, the most decisive parameters for the use of wet-handled coal ashes in construction are $SiO_2+Al_2O_3+Fe_2O_3$, loss of ignition (LOI) and moisture. Apart from that homogeneity in physical properties, density, water to cement demand, fineness and strength index (Adinugroho et al., 2022). According to ASTM C618, fly ash is classified into two classes: "C" and "F". In fly ash, the "Class F" amount of SiO_2, Al_2O_3 and Fe_2O_3 exceeds 70 %, whereas in "Class C", the amount of SiO_2, Al_2O_3 and Fe_2O_3 varies between 50 % and 70 %. "Class F" fly ash is a typical pozzolan, mainly consisting of silicate glass supplemented with iron and aluminum. "Class F" fly ash is mostly amorphous but also contains crystalline phases such as quartz, mullite, iron oxides, lime, and periclase. Usually, the crystalline constituents of fly ash are non-reactive. The amount of reactive SiO_2 must be between 40 and 50 % of the total mass, and the reactive Al_2O_3 amount is essential. The Ukrainian State Standard DSTU B V.2.7-128:2006 foresees technical parameters, which should be met for technogenic pozzolanic materials to be used in cement as pozzolanic materials (Table 1).

Table 1

Technical requirements to technogenic pozzolanic materials

No.	Parameter	Requirement
1	Student's *t*-test for one day ccompressive strength test	> 2.07
2	Finish Setting time, days	< 7
3	Water resistance at exposure sample in water more than 3 days	No mass loss, shape of tested sample is secured
4	Extension cylinder samples after 15 days, mm	< 15
5	SiO_2 reactive content, mass. %	> 25
6	Free lime, mass. %	< 2.5
7	Loss of ignition, %	< 5.0
8	Na_2O+K_2O content as Na_2O, mass. %	< 3.0
9	Cl^- content, mass. %	< 0.05

Since the CaO content in "Class F" fly ash is less than 10 %, the formation of calcium silicate hydrate (CSH) through the pozzolanic reaction requires a cement which, during hydration, forms portlandite. Consequently, the effectiveness of fly ash in concrete is mainly determined by its chemical components. Although the technical standards are available for rational usage of materials it would be required to consider the verification process of the standards in future explorations and tests.

Being stored in lagoons over the years, wet-handled coal bottom ash is well available both from volumes and seasonality point of view while PFA volumes are limited and mainly available in coal season when the cement market is low. At the same time providing proper process preparation measures (drying, grinding, grinding) it is possible to reach the same or similar properties as pulverized fly ash.

In the study (Tirkeş, 2021), an extensive tests program to compare the performance of PFA and Wet Bottom ash in Portland cement based mortars is presented. In the study, both types of ashes were samples at the same thermal power plant and obtained after firing the same type of coal. The test Program intended to use both types of ashes not only as supplementary cementitious materials but also as fine aggregate. Both types of ashes proved to be similar in terms of mineralogy and chemical composition. The main crystalline structures of both types of ashes were composed of quartz and anorthite, sillimanite. To conclude, the results showed that industrial by-products could be used in cement technologies at feasible amounts. Thus, this study helps the cement industry develop mortar technologies that have technical, economic, and ecological advantages for sustainable development like normal strength, durability, and environmental friendliness.

Authors (Cheeratot & Jaturapitakkul, 2004) proved that after moisturizing pulverized fly ash with high CaO content, the use as pozzolanic material in concrete is not efficient because the hydration process occurs. The use of such ash requires additional processing such as drying and milling that makes it economically infeasible, also reactivity of such fly ash is reduced.

Authors (Saengsov, Nguyen, Chatchawan & Tangtermsirikul, 2016) researched impact of replacement Portland cement with wet low-calcium pulverized fly ash at amount of 30 % and 50 % in concrete. It is demonstrated that concrete compressive strength based on wet pulverized fly ash increases in comparison to dry fly ash. Water in wet ash is the first dose of water in double mixing, which leads to an improvement in the interfacial transition zone of concrete made with wet ash.

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One option to overcome the problems associated with ordinary Portland cement (the 3H problems – high pollution, high cost and high energy consumption) (Zheng & Wu, 2021), is the development of alternative binders, among which alkali-activated binders (AAB) are promising. AAB production uses industrial wastes such as fly ash, rice husk ash and slag as precursors (Liu et al., 2022; Krivenko et al., 2019). The authors considered the possibility of using ash as a raw material for AAB (Deraman et al., 2015).

Additional milling (Akmalaiuly, Berdikul, Pundienė & Pranckevičienė, 2023), called mechanical activation, improves the reactivity of fly ash particles, makes fly ash material more uniform in terms of quality compared to raw fly ash. Ultrafine particles with an average size less than 10 µm or a specific surface area of more than 700 m^2/kg exhibit improved performance over a comparable control mixture. The replacement of cement by mechanically activated fly ash resulted in 20–30 % higher compressive strength at 7, 14, and 28 days than the replacement of cement by untreated fly ash.

At hydrothermal conditions when fly ah enters into a reaction with limestone, the main products of the reaction are α'L-C_2S, CaO, $C_{12}A_7$, the ratio among the products depends on temperature and time of reaction (Mazouzi, Kacimi, Cyr & Clastres, 2014). An advantage of fly ash-bellite cement in comparison to conventional Portland cement is the use of solid wastes, low burning temperature and lower production cost. However low strength of cement is a bottleneck for further development. Compressive and flexure strength of cement obtained at 850 °C at 28 days made up 19 MPa and 5.7 MPa correspondently (Guerrero, Goñi, Campillo & Moragues, 2004).

Fly ash can be used for the construction of the subgrade, backfilling of embankments and soil stabilization as a slow-setting, self-binding or active hydraulic additive (Novytskyi, Yatsenko &Topylko,

2022). Wet-handled coal ashes are used for the construction of subgrade and subbase of roads or low activity hydraulic additive in combination with cement for strengthening the soil base of III-IV road categories (Mozghovyi et al., 2014). Fly ash is widely used to stabilize expansive clays and natural soils with elevated alkali concentrations (Khan et al., 2022).

Conclusions

Nowadays wet-handled coal bottom ash is not extensively used as cementitious mainly due to its high moisture and coarse particles. At the same time providing that proper technological steps are applied (drying, milling or even burning) it is possible to achieve the same or similar performance as with pulverized fly ash. Considering the enormous available volumes of wet-handled coal bottom ash (360 million tonnes) in Ukraine it represents a high potential for cementitious usage in binding materials. Fly ash-based materials proved to be reliable cementitious both in cement and concrete.

Although technical standards on requirements for technogenic pozzolanic materials are in place in Ukraine it is advisable to consider verification of requirements for wet-handled coal bottom ash and make selection of specific requirements for wet-handled coal bottom ash to use as cementitious material. That requires significant scope of testing and research being done in the field.

Considering upcoming challenges with the availability of conventional cementitious such as GGBFS and the need to minimize using fossil pozzolanic materials investigations are to be considered in several directions: revision and verification of current technical requirements to technogenic pozzolan materials, selection of specific requirements for wet-handled coal bottom ash to use as cementitious material, studying possible technologies (grinding, drying, burning) to apply to bring parameters to required one, economic feasibility study for technologies, for specific wet-handled coal bottom ash source opportunities to be identified.

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МОЖЛИВОСТІ ВИКОРИСТАННЯ ЗОЛОШЛАКОВИХ ПРОДУКТІВ У В’ЯЖУЧИХ МАТЕРІАЛАХ: ОГЛЯД

Портландцемент залишається основним в’яжучим для забезпечення потреб зведення будівель та будівництва сучасної інфраструктури у всьому світі. Проте його виробництво є високоенергоємним та високоемісійним процесом, на який припадає понад 5 % світових викидів CO_2. Індустрія сталого будівництва зосереджена на впровадженні нових екологічно чистих рішень, пов’язаних із заміною традиційних матеріалів із високим вуглецевим слідом. Зменшення вмісту клінкеру в портландцементі стає першочерговим завданням, що відображено в дорожніх картах сталого розвитку цементної галузі європейських країн. Цей факт спонукає до пошуків і дій у різних напрямах, таких як удосконалення технологій помелу, розроблення хімічних добавок і розширення видів цементів із підвищеним вмістом мінеральних добавок із можливістю використання продуктів техногенного походження. Традиційною гідравлічною мінеральною добавкою у портландцементі є мелений гранульований доменний шлак, який є побічним продуктом виробництва сталі. Невдовзі обсягу доменного шлаку буде недостатньо, щоб задовольнити потребу будівельної галузі у мінеральних добавках, що спричинено зростанням попиту на шлак та зменшенням його кількості внаслідок зупинки деяких сталеливарних заводів, що використовують технологію з інтенсивним викидом CO_2. Тому розширення асортименту мінеральних добавок пуцоланічної дії, які є відходами промисловості, є першочерговим завданням для вирішення проблем цементної галузі, а також екологічних проблем їх зберігання. Все частіше в будівельних технологіях використовують матеріали, які порівняно недавно не становили цінності як мінеральні добавки через наявність легших варіантів. У статті розглянуто аспекти використання золошлакових матеріалів, яких в Україні на друге півріччя 2019 р. накопичилося у відвалах ТЕС близько 360 млн т, як мінеральної добавки для одержання в’яжучих.

Ключові слова: додатковий цементуючий матеріал; золошлаковий матеріал; зола-винесення; портландцемент; викиди CO_2; низьковуглецева в’яжуча речовина.