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ENVIRONMENTAL ASSESSMENT OF WET-HANDLED COAL BOTTOM ASH APPLICATION FOR CEMENT PRODUCTION

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The production of Portland cements with a reduced clinker content corresponds to the strategy of decarbonization of building materials, aimed at reducing the negative impact on the environment and climate changes. This direction of development of the cement industry requires the use of more mineral additives. The application of waste-derived materials as the main non-clinker component is promising. The advantage of using wet-handled coal bottom ash is its pozzolanic effect and high grindability, but the main disadvantage is increased moisture. The introduction of wet-handled coal bottom ash in the cement industry requires an integrated approach that covers aspects of technology, economics, and ecology. The article investigates the feasibility of using wet-handled coal bottom ash as an additive to binders in terms of environmental performance while ensuring the required strength indicators of Portland cement.

Keywords: Portland cement, wet-handled coal bottom ash, environmental performance, CO_2-emission, specific heat energy consumption, specific electricity consumption.

Introduction

Portland cement is a key construction material in order to satisfy the demands of building industry and civil infrastructure development. At the same time cement manufacturing process impacts significantly environment due to mining of large volumes of fossil raw materials, handling of raw materials and finish products, use of fossil fuels, calcination of carbonates and electricity with contribution to all world greenhouse emissions at level 5-8% (Cheng et al., 2023; Sroda, 2020). Increasing global urbanization is driving a steady trend towards expanding cement use and production, with an average annual growth rate of 4.1%, reaching four billion metric tons of cement annually, leading to an increase in global greenhouse gas emissions, which are expected to reach 1.58 Gt CO_2 in 2023 (Niu et al., 2024). Reducing carbon emissions from cement production is crucial to achieving the global climate goals set out in the Paris Agreement.

Efficient models for low-carbon development of the cement industry are country tailored, but common measures have been identified by the International Energy Agency (International Energy Agency, 2009): thermal and electrical energy efficiency, use of alternative fuels, replacement of Portland cement clinker, and carbon capture and storage (CCS).

New low-clinker cement types include the development of alternatives to cement as the main building material, but the production of the required quantity of such binders in the near future is problematic (Sroda, 2020). Today, the path to producing low-emission binders by replacing carbon-intensive clinker, which is an intermediate product in cement production, with other low-carbon materials – inert or reactive with cementing properties – remains the simplest from a technological point of view (Pamenter & Myers, 2021). This has led to a decrease in the clinker-cement ratio, which in 2017 was 77% in Europe (Sroda, 2020). According to the scenarios for decarbonising the German cement industry (Decarbonizing Cement and Concrete: A CO_2 Roadmap for the German cement industry, 2020), by 2050 the projected clinker-cement ratio will be 63% in the ambitious baseline scenario and 53% in the climate neutral scenario. At the legislative level, a significant reduction in the percentage of clinker in cement is considered in the EN 197-5 standard for the production of two types of cement - Portland-composite cement CEM II/C-M with a clinker content of up to 50% by mass and composite cement CEM VI with a clinker content of up to 35% by mass (Batog, Bakalarz, Synowiec & Dziuk, 2022).

Alternative materials to replace Portland cement clinker have most often been granulated blast furnace slag or coal-fired power plant fly ash. As evidenced by data (Shah, Miller, Jiang & Myers, 2022) their availability is decreasing. The stability of the clinker factor at 0.75 since the beginning of the century demonstrates the increasing use of other cementitious materials to replace clinker, in particular limestone, zeolite, oil shale (Batog et al., 2022; Sanytsky, Kropyvnytska, Fic & Ivashchyshyn, 2020). Such mineral cementitious account for 21% of all substitutes, and non-traditional substitutes such as calcined clay, silica, and pozzolanic materials from industrial waste are also being investigated (Scrivener, John & Gartner, 2018). A limitation of the use of metakaolin as a clinker substitute is the need for heat treatment (at temperatures of 700–850 °C) to achieve the targeted reactivity, which influences on environment negatively, but this impact is smaller compared to clinker production (Ascensão, Farinini, Ferreira & Leardi, 2024).

Sustainable Portland cement production is supported by expanding the range of suitable materials. CO_2-efficient raw materials are usually various waste-derived materials. The advantages of using them are the absence of quarries development, reduced landfill use, and saving of natural raw materials. In 2023, EN 197-6:2023 Cement with recycled building materials was adopted, using recycled concrete fines in an amount of 6-35 wt.% to obtain CEM II/A-F and CEM II/B-F Portland cement or as a component of composite-Portland cement of the second type and composite cements of the fifth type. The authors (Sobol et al., 2020; Hunyak, Hidei, Sobol & Petrovska, 2023) showed the use of ash from paper production waste as a cementing material for binders.

One of the investigated materials that can be used to replace Portland cement clinker is wet handled coal ash from thermal power plants. According to the Ukrainian European Business Association (European Business Association, 2021), the volume of wet handled coal ash in Ukraine will be approximately 415 million tons by 2035. Such ash is not widely used mainly due to its high moisture content (usually above 20%) and the presence of coarse particles, which requires additional technological preparation before use as an additive to cement (Cheeratot & Jaturapitakkul, 2004; Sobol & Marushchak, 2024; Tirkeş, 2021). In addition, one of the most key quality control parameters in wet handled coal ashes is unburnt carbon content, which is analysed as loss of ignition, it is necessary to assure that unburned carbon does not impair overall quality of binder. At the same time, the use of ash does not lead to a deterioration in the water consumption of the binder and often improves the rheological properties of concretes based on such binder. In addition, the ash is characterized by pozzolanic properties, which contribute to the strength formation of Portland cement when used in optimum quantities (Permatasari, Sodri & Agustina, 2023). The introduction of an increased amount of ash initially causes a decrease in the strength of Portland cement at an early age and the strength development, which requires the use of certain technological measures, in particular, increasing the fineness of the binder, the use of plasticizers, hardening accelerators in the concrete manufacturing (Sanytsky, Kropyvnytska, & Hevyuk, 2021).

However, a comprehensive assessment of the use of wet handled coal ash, in addition to compliance with standard criteria, i.e. mechanical (compressive and bending strength), durability (frost resistance and chemical resistance), as well as economic, should include an analysis of its life cycle (Ondova & Stevulova, 2013; Permatasari, Sodri & Agustina, 2023)

Life cycle assessment (LCA) is widely used in research around the world as a tool for assessing environmental impacts. There are various models for performing LCA, such as process-based LCA, economic input-output LCA (EIO-LCA), triple outcome LCA (TBL-EIO), hybrid LCA, extended LCA with attribution (AALCA), and social LCA (S-LCA) (Tait & Cheung, 2016).

The international database of building materials Ecoinvent uses for comparison and assessment of such environmental parameters as primary energy consumption, global warming potential (CO_2 absorbed, eq.), environmental acidification potential (solid emissions of SO_2, eq.), ground-level ozone formation potential, ozone depletion potential - ODP, and ecological eutrophication potential. Studies based on the methodological standards ISO 14040:2006 and ISO 14044:2006, considering three impact categories, such as primary energy demand, water demand and global warming potential, prove that using waste derived materials instead of limited natural resources can significantly reduce the environmental impact of construction products (Bribián et al., 2011).

The purpose of the research is to study the impact on the environment when using wet-handled coal ash from thermal power plants as an active mineral cementitious in the manufacture of binders.

Materials and Methods

In the study, the wet-handled bottom coal ash from power stations is considered as cementitious material at Portland cement manufacturing for the two scenarios. In the first scenario (Scenario 1) the calculation is conducted for 10% substitution of granulated blast furnace slag (GBFS) with wet-handled bottom coal ash without replacing Portland cement clinker to obtain cement CEM II/B-M. In the second Scenario (Scenario 2), the calculation is conducted for the substitution of 10% Portland cement clinker with wet-handled bottom coal ash for cement with GBFS to obtain cement CEM II/C-M.

To assess the possibility of wet-handled ash use from power stations in cement manufacturing the following key performance indicators were used:

- specific heat consumption for binder manufacturing;
- specific electricity consumption for binder manufacturing;
- specific greenhouse gas emissions as CO_2 at binder manufacturing.

As reference binder Portland cement CEM II/B-S 42.5R is considered, this type of cement is available in the domestic Ukrainian market. The approximate analysed content and main technical parameters are given in Table 1. It is intended to keep standard compressive strength at the level of reference binder, namely 50 MPa, which defines the most stringent conditions of processing binder. In addition, it is important to note that obtaining binders with lower standard compressive strength is easier and lower energy consumption from the perspective of manufacturing process.

Table 1

Approximate analysed content and main technical parameters of reference binder

Approximate analysed content, %			Specific surface, $cm^{2/}g$	Sieve residue at 32μм, %	Compressive strength at days, MPa	
Portland cement Clinker	GBFS	Gypsum			2	28
70	25	5	5000	5	27	52

The heat for clinker manufacturing and heat required for preparation (drying) of GBFS and wet-handled bottom coal ash are taken into account for the calculation of specific heat consumption for manufacturing of binder. Specific heat consumption for clinker manufacturing is considered at level 3200 MJ/t for the dry process with calciner and preheater (Schorcht, Kourti, Scalet, Roudier & Sancho, 2013). In case another process for clinker manufacturing is used, the impact from the cementitious material addition into binder increases due to higher specific heat requirements for clinker production, for example for the wet method it is 5000 – 6400 MJ/t.

Cementitious materials contain natural moisture, which shall be evaporated at a separate grinding process. For this purpose, the milling equipment hot gas generators are applied with the firing of different fuels, for example, natural gas, coal, and alternative fuels based on agricultural wastes. Using waste gases from clinker manufacturing is more efficient. In the wet process technical possibility of using waste gases from the clinker manufacturing process is limited. Assessment of the impact of wet-handled bottom coal ash use as cementitious material is conducted for both hot gases from hot gas generators and waste gases from the clinker manufacturing process (Fig. 1). The moisture of GBFS of 8% and moisture of wet-handled bottom coal ash of 20% are taken into account. Heat requirement for drying of cementitious material is considered at a level of 943 kcal/kg moisture (Locher, 2006).

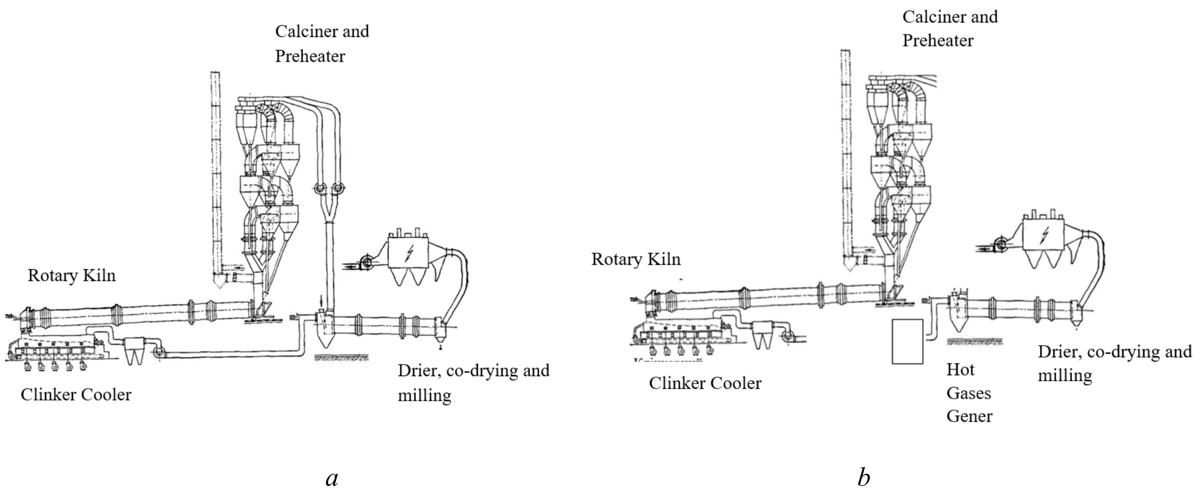

Fig. 1. Possibilities to use hot gases for cementitious drying:
a) waste gases from pre-heater or clinker cooler, b) using additional fuels in hot gases generator

The main goal for different cementitious using or Portland cement clinker content reduction in binder is the achievement of equivalent qualitative properties of binder in comparison to reference binder. The main parameter of quality of binder apart from compressive strength at 28 days is early compressive strength usually tested after 2 days since the speed of hydration of binder defines the construction dynamic and efficiency of assets use. Therefore, achievement of early and standard strength was considered as the main pre-condition during assessment.

The speed of pozzolanic reactions increases with the time of hydration and most mineral cementitious materials contributes to compressive strength at later periods, usually after 7 days. At the same time contribution of mineral cementitious materials into early compressive strength is insignificant since it is necessary to ensure a sufficient volume of calcium hydroxide, formed during Portland cement clinker hydration.

Due to this fact, wet-handled coal ash content in binder for GBFS substitution is considered at a level of 10% for a conservative approach, assuring the possibility of combining $Ca(OH)_2$ with the active part of SiO_2 of cementitious material. In case of high quantities of wet-handled bottom coal ash introduced into binder, the volume of calcium hydroxide could be insufficient and cementitious material can become inert in standard terms of testing (28 days) but in the later ages, thanks to ongoing pozzolanic reaction compressive strength continues to increase. To intensify pozzolanic reactions it is necessary to increase the amount of calcium hydroxide in the hydration system, which is possible to achieve with increasing lime saturation factor of Portland cement clinker, the introduction of lime or calcium hydroxide, and an increase of clinker particles fineness. As a completely using optimization processes it is possible to achieve 20% or more wet-handled bottom coal ash content in binders.

In the current assessment, it is assumed that the contribution to early compressive strength is made only by Portland cement clinker. Achievement of required early compressive strength with reduction of clinker content is possible by applying the following techniques: (1) increase fineness of Portland cement clinker particles, (2) using strength enhancers or both. The current assessment is conducted considering only Portland cement clinker fineness increase.

Specific electricity consumption for binder manufacturing is calculated as the sum of specific electricity consumption for grinding each component of binder (Portland cement clinker, gypsum, GBFS, wet-handled bottom coal ash) to achieve the required fineness as well as specific electricity consumption of clinker manufacturing process. Specific electricity requirements for grinding components of binders are given in Table 2 (Holderbank Engineering Book, 2000). Specific electricity consumption for clinker manufacturing is taken at a level of 69.3 kWh/t considering specific electricity consumption for cement manufacturing of

110 kWh/t cement and the fact that all processes required for clinker manufacturing make up 63% of total electricity consumption for cement (mining and grinding raw materials, burning and clinker cooling, coal grinding) (Atmaca & Atmaca, 2016).

Table 2

Specific electricity consumption for grinding of different materials

Material	Specific electricity consumption	
	Specific surface 5000 cm^2/g	Specific surface 5800 cm^2/g
Portland cement clinker	38	44
GBFS	44	-
Wet-handled bottom ash	15	-
Gypsum*	38	-

*Considered as for Portland cement clinker.

Calculation of specific greenhouse gas (CO_2) emissions is conducted for direct and indirect emissions. Direct emissions cover greenhouse gas emissions from the manufacturing process and are part of Scope 1 emissions. Indirect emissions of greenhouse gases (CO_2) are connected to emissions from electricity production (Scope 2) and emissions connected mainly to the manufacturing processes and transportation of raw materials (Scope 3).

Direct emissions of greenhouse gases for binder manufacturing are calculated for the use of hot gases for drying cementitious material for both using waste gases from the kiln system and using additional fuel for cementitious drying. For Portland cement clinker the value 0.83 t CO_2/t clinker is used (Guidance Document on CBAM Implementation for installation operators outside the EU, 2023), which corresponds to the level of emissions after dry process kiln with calciner and preheater without the use of calcined raw materials and alternative fuels containing biomass. In the case of using additional fuel for cementitious drying emission factor of 55.7 t CO_2/TJ is applied (Greenhouse gas emission factors and net calorific values (NCVs) of fuels per unit mass, 2023). An emission factor of 226 t CO_2/GWh is applied for the calculation of indirect emissions of CO_2 after electricity production (Energy profiles. Ukraine, 2024).

Emission factors of 0.083 t CO_2/t and 0.008 t CO_2/t are considered for assessment of indirect emissions of CO_2 after generation and transportation of GBFS and wet-handled bottom coal ash correspondingly. Gypsum emission factor for ground limestone (0.032 t CO_2/t) is considered since both materials are similar in their physical properties (Hammond & Jones, 2011).

Results and Discussions

When using waste heat from kiln exit gases for drying cementitious materials specific heat consumption for manufacturing of binder when substituting the GBFS with wet handled coal ash (Scenario 1) is the same as in reference binder CEM II/B-S 42.5R and lower by 135 MJ/t due to decreased clinker content in binder according to Scenario 2, despite higher wet handled ash moisture than GBFS (Fig. 2). When using hot gases from hot gas generator for drying cementitious materials specific heat consumption for Scenario 1 is higher than for reference binder by 64 MJ/t due to higher moisture of wet handled coal ash. At the same time, specific heat requirements for binder manufacturing in the case of Scenario 2 were reduced by 1.1 times (by 221 MJ/t) compared to reference binder due to the reduction of Portland cement clinker content, which is characterized by high energy intensity.

Calculated specific electricity consumption makes up 0.103 MWh/t for reference binder (Fig. 3). At the assessment of specific electricity consumption for binder at Scenario 1 clinker content reduction was not envisaged and the expected specific surface was at the same level as for reference binder – 5000 cm^2/g. Specific electricity consumption for binder at Scenario 1 is lower by 3.0 % because of the better grindability of fly ash in comparison to GBFS.

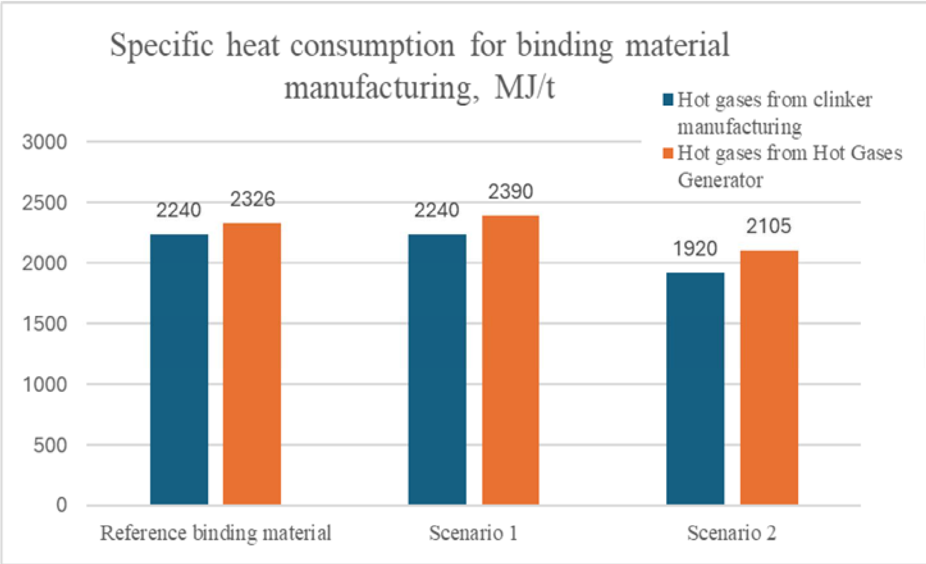

Fig. 2. Specific heat consumption for binders' production

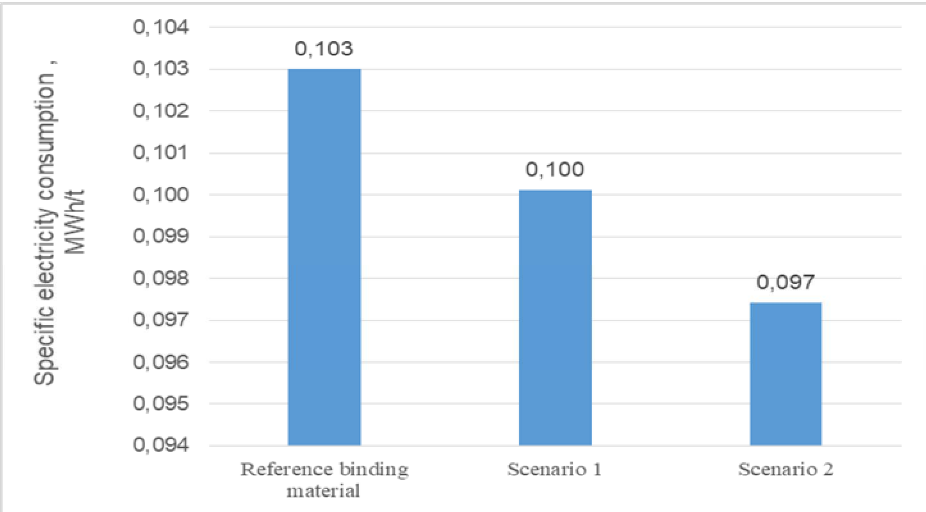

Fig. 3. Specific electricity consumption for binders' production

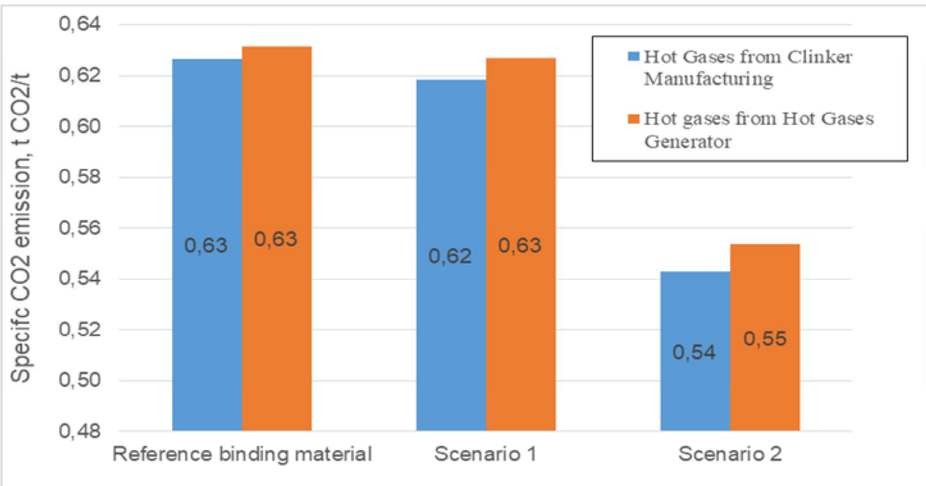

Fig. 4. Specific greenhouse gases emissions at binders production

Obtaining of binder in Scenario 2 envisages the reduction of Portland Cement Clinker content therefore for the achievement of required compressive early strength it is necessary to grind clinker finer, at the same time fineness of cementitious material remains the same as in reference binder (5000 cm^2/g). The approximate specific surface of clinker particles in Scenario 2 makes up 5850 cm^2/g) and binder approximately 5510 cm^2/g. Considering electricity consumption for additional clinker grinding and its replacement with wet-handled bottom ash with better grindability, the estimated specific electricity consumption at Scenario 2 makes up 0.097 MWh/t, that is by 5.8% less in comparison to reference binder.

The values of specific greenhouse gas emissions (in terms of CO_2 – CO_2eq) for manufacturing of reference binder and binder in Scenario 1 are not very different and make up approximately 0.63 t CO_2/t (Fig. 4).

At the same time for binder in Scenario 2 specific greenhouse gases emissions (CO_2eq) reduced by 14.3 % that corresponds to 90 kg CO_2eq in the case of using waste heat gases from clinker manufacturing by dry process and by 12.6 % in the case of using heat of hot gases from the hot gas generator.

Conclusions

Portland cement is a material requiring high energy uses and a significant carbon footprint for its manufacturing. Maintaining low clinker content in binders is key to its low energy intensity and sustainability. Wet-handled bottom coal ash has high grindability, at optimum level in binder guarantees pozzolanic reaction during the hydration process and does not require significant energy consumption to reach required properties. The main disadvantage of wet-handled bottom coal ash is its high moisture which requires additional heat for evaporation. However with the limited availability of GBFS in the future, using the wet-handled bottom coal ash could be justified in general. Using wet-handled bottom coal ash for partial substitution of GBFS or clinker is appropriate for both energy consumption and greenhouse gas emissions. According to the conducted assessment partial substitution of GBFS leads to a similar level of greenhouse gas emissions in comparison to using GBFS only (0.63 t CO_2/t), at the same time replacement of clinker, the level of greenhouse gas emissions makes up 0.55 t CO_2/t of binder.

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ЕКОЛОГІЧНА ОЦІНКА ЗАСТОСУВАННЯ ЗОЛОШЛАКОВИХ ПРОДУКТІВ ДЛЯ ВИРОБНИЦТВА ЦЕМЕНТУ

Портландцемент є одним з найбільш енергоємних матеріалів із значним вуглецевим слідом його виробництва. Курс на зниження вмісту портландцементного клінкеру у в’яжучих є запорукою отримання стійких екологічних матеріалів. Найпоширенішою мінеральною добавкою при виготовленні в’яжучих був доменний гранульований шлак, проте при прогнозованій обмеженій його доступності в майбутньому виникає необхідність розширення сировинної бази неклінкерних компонентів. З точки зору зниження впливу на довкілля практичний інтерес представляє використання відходів. Вугільна зола мокрого видалення характеризується високою розмелювальною здатністю, при введенні у в’яжучі в оптимальній кількості забезпечує протікання пуцоланових реакцій при їх гідратації, не потребує значних енергетичних затрат для досягнення їх необхідних властивостей. Основним недоліком вугільної золи є її висока вологість, що вимагає додаткових енергетичних затрат на сушіння. Для сушіння матеріалів з високою вологістю перспективним є використання теплоти відхідних газів, утворених при сухому способі виробництва портландцементного клінкеру. Згідно з проведеними розрахунками використання вугільної золи як для часткової заміни доменного гранульованого шлаку у в’яжучому, так і для заміни портландцементного клінкеру є доцільним з точки зору питомої витрати енергоносіїв та викидів вуглекислого газу. В’яжуче з частковою заміною доменного гранульованого шлаку на вугільну золу характеризується таким самим питомим показником викидів вуглекислого газу, як і при використанні тільки доменного гранульованого шлаку (0,63 т CO_2/т в’яжучого), в той же час при заміні портландцементного клінкеру на вугільну золу показник викидів вуглекислого газу становить 0,55 т CO_2/т в’яжучого. Результати цього дослідження можуть дати розуміння для розроблення політики підвищення стійкості випуску в’яжучих з використанням різних типів мінеральних добавок.

Ключові слова: портландцемент, золошлаковий матеріал, екологічний показник, викиди CO_2, питомі витрати теплової енергії, питомі витрати електричної енергії.