REDUCING THE CARBON DIOXIDE FOOTPRINT OF INORGANIC BINDERS INDUSTRY

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Abstract

Reducing the carbon footprint is a strategic objective established for environmental protection the European Union. It is proposed that Europe be climate neutral until 2050. This objective can also be achieved in Romania, if the large energy-consuming industries, that produce inorganic binders (cement, lime, plaster), implement the best available techniques. These techniques must ensure climate neutrality throughout the value chain of construction materials, such as: (1) reducing direct and indirect CO_2 emissions; (2) increasing the use of alternative material and energy resources; (3) developing products with a reduced carbon footprint; (4) contributing to society's adaptation and combating climate change; (5) carbon capture, usage and storage. Partial substitution of the natural fuels with the alternative fuel and energy resources from sorted industrial and municipal waste can be promising techniques for the reducing of the carbon footprint. Thus, in the combustion process the following wastes can be used: industrial waste (oil tankers, used tires, non-recyclable packaging), non-recyclable municipal waste (wood, paper, plastic, textile, biomass, etc.), agricultural wastes, etc. This paper presents a case study regarding the reduction of the carbon dioxide footprint by using alternative resources such as tires waste and in the cement manufacturing at Tașca Cement Plant from Neamț country (Romania). It has been proven that the use of these resources leads to sustainable benefits related to the partial conservation of non-renewable energy and material resources, as well as to the reduction of carbon dioxide emissions. The measurements CO_2 content at the chimney of the clinker showed a decrease between 0.4 % to 3 %. This proves that the introduction of slag at the cold area of the kiln has a positive effect on the reduction of CO_2 emissions and implicitly over the environment.

Keywords: *alternative fuels, alternative materials, cement production, emission, slag, tires waste*

1. INTRODUCTION

Cement is one of the indispensable materials used in civil and industrial construction, as well as in infrastructure, but it is a major consumer of energy and raw materials [1, 2, 3, 4].

The cement industry is one of the largest industrial sources of CO_2 emissions in the world, accounting for 2.2 Gt/year in 2018, or more than 6 % of global emissions from fossil fuel use [4, 5, 6]. Over the years, the cement industry has substantially reduced CO_2 emissions per ton of cement by improving energy efficiency, replacing fossil fuels with waste that can sometimes be considered "carbon neutral" and increasing the use of additives in the cement product [5, 7]. The scope for further reductions by these means is becoming limited, but there is a growing need to reduce CO_2 emissions to avoid climate change [7, 8]. More than 60 % of total CO_2 emissions from a modern cement plant come from the thermal decomposition of raw materials and this CO_2 cannot be avoided by using alternative energy sources, only by finding and using alternative raw materials or new methods of capturing, transporting, storing and using CO_2 emissions [4, 5, 9].

There are no legislative limits on CO_2 emissions, only commitments by major cement producers, especially those in Europe. Major players in the global cement and concrete industry have announced that they want to "accelerate the transition" to cleaner production with a further 25 % reduction in CO_2 emissions by 2030, and to reach their goal of CO_2 neutrality in 2050 [4, 10, 11]. Companies in the sector are committed to "avoiding five billion tons of CO_2 emissions by 2030", equivalent to "15 billion commercial flights between Paris and New York", the Global Cement and Concrete Association (GCCA) announced in a statement [12].

The main stage in cement production is the production of clinker, with about 1.6 tons of raw materials needed to produce one ton of clinker. The clinkering of the raw material mixture is the most energy-intensive technological operation in the Portland cement production process [9, 10, 11].

Any method of reducing the energy required has a beneficial influence on both the viability of the cement plant and the environment [5, 13]. One of the approaches to reduce thermal energy consumption is the partial substitution of traditional raw materials (limestone, clay, gypsum) with alternative, fluxing materials, among which by-products of the steel industry such as blast furnace and steel mill slag [10, 14]. Another approach is the use of alternative fuels [10, 15].

Nowadays, the use of combustible waste in some cement plants in Europe contributes at least 50 % of the total heat requirement used in the cement clinker manufacturing process [4, 5, 13]. By burning the waste in the clinker kiln, all the gaseous acidic compounds formed by combustion are neutralized by the basic nature of the raw material used to make the clinker and then incorporated into the clinker. The interaction in the clinker furnace between the flue gas and the raw material results in a complete removal of the non-combustible material from the alternative sources and ensures its incorporation into the clinker. By burning alternative fuels rather than conventional fossil fuels such as coal, less CO_2 is removed, which results from the lower carbon content of alternative energy sources [5, 13].

Such alternative fuels typically include plastics and paper/paper from commercial and industrial activities (e.g. packaging waste or manufacturing waste), waste tires, waste oils, biomass waste (e.g. straw, untreated waste wood, dry sewage sludge), textile waste, residues from car dismantling operations (automatic residue shredder – ASR), hazardous industrial waste (e.g. certain industrial sludges, impregnated sawdust, spent solvents) as well as obsolete pesticides, chemicals and pharmaceuticals [6, 11, 13].

Metallurgical slags are technical silicates resulting from thermal processes, as melts during the extraction of metals from ores, as well as from metal melting [11, 16]. They fall into two classes: (1) blast furnace slag (granulated slag and non-granulated slag), (2) steel slag (oxygen-based slag, electric steel slag and secondary slag). The chemical and implicit mineralogical composition of the slags is variable, finding all the specific oxidic components of Portland cement CO_2 measurements at the chimney of the clinker plant showed a decrease in CO_2 concentration in the exhaust gas.

In some European countries, Australia and the USA, the use of blast furnace slag is also practiced in the production of Portland cement clinker [17, 18, 19]. Thus, the use of blast furnace slag, as a raw material to produce clinker, has been practiced since 2002 at a plant in Hungary, replacing limestone and clay in the proportion of 1 – 1.5 % [17]. Also, Adelaide Brighton Cement (Angaston Australia) annually uses approx. 50000 t blast furnace slag as raw material [18]. Because the slag has undergone a thermal process, less fuel is required for combustion in the furnace, thus reducing CO_2 emissions from combustion. In addition, the calcium oxide in the slag replaces some of the limestone in the raw mix, which is another source of CO_2. Since granulated slag does not require crushing, it indirectly reduces CO_2 emissions generated by electricity. Thus, the team from Angaston demonstrated that, in the case of using slag as a raw material, CO_2 emissions are reduced by approx. 9000 tons of CO_2/year [18].

According to EUROSLAG Statistics Report [20] at the level of 2016, in Europe, blast furnace slag production was 24,600,000 t, of which 78.9 % is granulated blast furnace slag. The largest share of blast furnace slag was used in cement/concrete production and in road construction. Thus, 81.4 % of the total amount of blast furnace slag was used for cement/concrete production and 20.9 % in road construction [21, 22, 23].

According to EUROSLAG Statistics Report [20] at the level of 2016, in Europe, the production of steelworks slag was 18.4 Mt, of which 56.6 % is BOF steelworks slag and 32.2 % EAF slag. The main area of use for steelworks slag was road construction, where 46 % of the total quantity was used. Other areas of use of steelworks slag were metallurgy with 15.3 %, civil engineering 2.2%, cement/concrete 4.4 %, fertilizer in agriculture 2%. an important part, approx. 23 %, was landfilled [21, 22, 23].

In the following, research from the literature is presented that refer to the possibility of using different types of slag (blast furnace and steel mill slag) in the raw material mix in cement production. Based on these considerations, the present work aims at comparative studies on the composition of flue gas resulting from the manufacture of Portland cement clinker using alternative fuels alongside conventional fuel and metallurgical slag as raw material. For the raw material mixture in the production of Portland cement clinker, the slag source found near the Taşca cement plant, i.e. black blast furnace slag from the ArcelorMitall Galați steel plant (Romania), is used.

2. MATERIALS AND METHODS

2.1. Materials

2.1.1. Alternative fuels

Studies on the possibilities of reducing the carbon footprint were carried out from 25 to 28 June 2021, using conventional fuel (a mixture of coal and petroleum coke) together with alternative fuels such as: used tyres, RDF, cellulose, shredded plastic for the combustion process. The fuels used, their quantities (in tons) and their net calorific value (NCV, J/g) are shown in Table 1.

Specification		**Fuels**				
		Coal mix	**Tires**	**RDF**	**Cellulose**	**Shredded plastic**
Test A	Quantity (t)	110.01	52.56	46.97	8.86	1.16
	Pci (J/g)	28022	25000	15859	20503	18066
Test B	Quantity (t)	110	52.22	42.42	14.15	
	Pci (J/g)	28320	25000	14250	12180	

Table 1. Used fuels (from the kiln operation sheets).

2.1.2. Air-cooled blast furnace slag

In view of the results obtained on a global laboratory scale and the quantities available, it was decided to carry out the industrial experiments using black slag from the Galati blast furnace. The slag was chemically analyzed in the laboratory of the Taşca cement plant. The results showed that the slag used has the following chemical composition: 36.5 % SiO_2, 42 %, CaO, 10 % Al_2O_3, 0.5 % Fe_2O_3 (expressed as average %wt values). The alkali content (0.81 – 1.39 %) is comparable to that of a kiln feed flour. The particle size analysis of the slag showed a high proportion of the coarse fraction above 25 mm (42.7 %). It should be noted that the literature recommends that the maximum size of the slag, in case of cold head kiln feeding, should be approx. 20 mm [20].

2.1.3. Raw materials used

The raw materials used to produce the experimental batch of raw meal were those used in the current production: limestone from Bicaz - Chei quarry (RO), marl Țepeșeni (RO), sand Văleni (RO) and pyrite ash (RO). The chemical composition of the raw materials used was analyzed in the laboratories of the plant from 25 to 28 June 2021 and is shown in Table 2 (expressed in average %wt values).

Period	Raw material	Composition, %wt							
		CaO	SiO_2	Al_2O_3	Fe_2O_3	MgO	K_2O	Na_2O	SO_3
25 – 27.06.2021	Limestone	52.39	3.87	1.22	0.55	0.83	0.26	0.21	0.42
	Marl	14.36	118.04	13.11	6.09	2.21	2.48	0.28	0,56
	Sand	-	87.8	-	-	-	-	-	-
	Pyrite ash	-	-	-	89.34	-	-	-	-

Table 2. Chemical composition of the raw materials used.

2.2. Methods

Industrial experiments have been carried out in which CO and CO_2 emissions have been monitored on the one hand, and on the other hand, the conditions to produce clinker have been determined in relation to the characteristics of the raw materials.

During the experimentation, CO_2 emission measurements were carried out at the stack of the clinkering plant. The hydraulic potential of the clinker plant was also evaluated, as well as the environmental impact of using slag as a raw material for clinker production.

The proposed conditions for the industrial experiments on the use of black blast furnace slag as raw material for cement clinker production are:

- introduction of slag directly into the kiln in the ascending column, using the feed system of shredded bark and rubber;
- quantity of slag introduced: about 3 t/h;
- use of a batch of flour with LSF approx. 103;
- obtaining a clinker with LSF = 98, MSi = 2,4, MAl = 1,4 and CaO_{free} max. 2 %.

Industrial experiments were carried out on the second manufacturing line from 25 to 28 June 2021. Data on the batches made for the experimental trials are shown in Table 3. The difference between the two tests was that in Table 3 the amount of flour fed into the kiln was reduced while maintaining the same amount of slag to determine the optimal conditions for its use with the aim of reducing CO_2 emissions as efficiently as possible.

Production stage		Date of the experiments	Quantity (t)
Raw meal		25.06.2021 time 15:00 – 27.06.2021 time 15:00	≈ 13500
Clinker	**Test A**	26.06.2021 time 12:00 – 27.06.2021 time 12:00	≈ 3150
	Test B	27.06.2021 time 12:00 – 28.06.2021 time 8:00	≈ 2550
Cement	**CEM I 42.5R**	27.06.2021 time 11:00 – 28.06.2021 time 3:00	≈ 2360

Table 3. Experimentation period, size of batches made.

2.3. Clinkerization parameters

The main parameters of the plant were monitored during the experimental period and are presented centrally in Table 4.

Process parameters	**UM**	**Experimental conditions**		**Current operation**
		Test A	**Test B**	(25.08.2020)
Operating time	h	24	20	24
Raw meal flow	t/h	205.5	200.2	205.8
Slag flow	t/h	3.3	3.3	-
Kiln RPM	rot/min	1.97	1.86	1.98
Max.temp kiln shell	°C	404	405	398

Table 4. Main parameters of the clinkerization process.

3. RESULTS

Cement production starts in quarries, with the excavation of limestone and clay. These are crushed and then transported to the plant by conveyor belt systems and/or rail. Limestone and clay contain large amounts of calcium carbonate and aluminum silicates.

The limestone and clay are finely ground together with other raw materials that provide iron and/or silicon, each of which is carefully dosed and analyzed to follow the recipe for preparing the flour, which is then fed into the kiln to make clinker. The raw flour, heated in the rotary kiln to a temperature of up to 1,450 ^{O}C, is transformed by flash cooling into a new, crystalline, granular material called Portland clinker, which is an intermediate - but essential - product in cement manufacture.

Grinding the clinker, together with well-controlled dosages of gypsum and manufacturing additives (slag, limestone, etc.) to a particularly high fineness, produces the final product - cement - which is stored in silos and delivered in bags or in bulk in specialized trucks. One of our objectives is to reduce the amount of natural raw materials used in the production processes, i.e. to protect - also by this measure - the environment. We use alternative raw materials based on waste or by-products from other industries in the production of clinker as well as in the grinding of cement, taking strict care that the final product fully complies with the requirements of the European product standard SR EN 197-1:2011 [24].

3.1. Preparing and characterizing the raw mixture

The conditions required to obtain the batch of flour used during the experiments are shown in Table 5. Variations in raw material dosage, modular and physical characteristics of the flour samples during the experimentation period, summarized in Table 6 (average values), compared to values related to current operation. The feed rate of the flour mill during the experimentation averaged 290 t/h compared to 287 t/h during the current period of operation. From the data presented comparatively for the quality parameters of the raw mix, the following was observed:

- a relatively large range of variation (95.8 – 110.8), but the average LSF value was 103 as proposed;
- mean values for MSi and MAl close to the proposed 2.53 and 1.69 respectively;
- more advanced grinding fineness of the flour obtained for the experimental batch compared to the one currently produced (18.37 % versus 19.92 %).

Raw material dosage (%)				**Modular features**			**Fineness (%)**	
Limestone	Marl	Sand	Pyrite ash	LSF	MSi	MAl	R020	R009
≈ 74.5	≈ 21.5	≈ 3	≈ 1	103	2.5	1.7	≈ 3	≈ 19

Table 5. Dosage of raw materials and modular and physical characteristics required for the raw mix

Characteristic	**Raw material dosage (%)**				**Modular features**			**Fineness (%)**	
	Limestone	Marl	Sand	Pyrite ash	LSF	MSi	MAl	R020	R009
Experimental conditions	74.42	21.53	3.16	0.89	103.08	2.53	1.69	3.17	18.37
Current operation	74.21	22.10	2.99	0.7	105.84	2.60	1.77	3.53	19.92

Table 6. Average values for dosage of raw materials, modular and physical characteristics of raw meal

3.2. Preparing and characterization of the clinker mixture

Table 7 shows the average value of the physical and modular characteristics of the kiln feed flour for the industrial experimental conditions compared to the current operation. Examination of the data presented in the table shows that:

- the mean value of LSF for test A was ca. 105 being comparative to that of raw meal corresponding to a current period of operation. The flour used in test period B had an LSF of 103.16 comparable to that proposed for the experiment;
- silica modulus and alumina modulus were similar in both tests: MSi = 2.6 and MAl = 1.6
- burnability (BF) is similar for the kiln-fed flour during the two tests, the BF index value being 115.5 for Test 1 and 115.7 for Test A.

Also, in Table 7 the chemical composition of the kiln feed flour from the period of the experiments (Test A, Test B) compared to the characteristics of the flour from the current production is shown. The oxidative composition of the oven feed flour from the experimental period is comparable to that of the raw meal in current use.

Characteristic		**Experimental conditions**		**Current operation**
		Test A	**Test B**	(25.08.2020)
Physical characteristics	R020 (%)	3.44	3.7	3.47
	R009 (%)	20.19	20.52	20.22
Modular characteristics	LSF	105.02	103.16	105.95
	MSi	2.62	2.60	2.61
	MAl	1.64	1.60	1.73
Chemical characteristics	% CaO	43.55	43.46	43.55
	% SiO_2	13.07	13.26	12.93
	% Al_2O_3	3.10	3.19	3.18
	% Fe_2O_3	1.89	1.92	1.79
	% SO_3	0.3	0.30	0.31
	% MgO	1.01	1.01	1.02
	% K_2O	0.65	0.65	0.69
	% Na_2O	0.13	0.13	0.13
	% Na_2Oechiv	0.56	0.56	0.58

Table 7. Physico-chemical characteristics for raw meal kiln feeding

4. DISCUSSION

4.1. Specific heat consumption

The total throughput of material fed into the kiln (flour + slag) was 208.8 t/h for Test A and 203.5 t/h for Test B. This difference in feed rates is because in Test B the flour feed to the kiln was reduced while the slag feed rate was kept constant. The kiln speed during Test A (1.97) was comparable to that recorded during current operation. A lower speed was observed during Test B.

From the data recorded in the technology sheet for the mantle temperature, a slight increase is observed during the experiments which can be attributed to a possible reduction in the thickness of the solder layer. Thus, the average value of the mantle temperature was higher by 60 ^{O}C respectively by 70 ^{O}C compared to the current operating period analyzed.

The specific heat consumption for the two tests was calculated based on the data recorded in the furnace operating records, considering 12 hours of continuous operation, as follows: for Test A – shift 2 on 26.06.2021 and for Test B – shift 2 on 27.06.2021. The calculation of the specific heat consumption was made considering:

- clinker production calculated as follows: total quantity of flour fed into the kiln divided by the transformation factor (Ft) to which 3.3 t slag is added for each hour of operation; Ft = 1.62.
- the quantity of fuels used and the lower heating value of the fuels in Table 1.

The specific consumption values are given in Table 8. The results show a reduction in specific heat consumption of approx. 4 % for test A and by approx. 4.5 % for test B, compared to the current operating period analyzed.

The decrease in specific heat consumption is mainly due to the reduction in the amount of fuel used in the clinker plant as well as to the presence of slag which has already undergone a thermal transformation and is mainly composed of silicates and aluminosilicates. The calculated hourly clinker production was 130.47 t/h during test 1 and 127.63 t/h during test B. For the current operating period the hourly clinker production was 127.16 t/h.

Indicator	**UM**	**Experimental conditions**		**Current operation**
		Test A	**Test B**	(25.08.2020)
Specific heat consumption	GJ/t	3.414	3.394	3.556

Table 8. Specific heat consumption

4.2. Emissions measured at the chimney of the cement plant

Data on CO_2, CO, NO_x, SO_2, HCl, CO, HF and dust (PM) emissions were processed from continuous monitoring of the plant during the experiments. The information obtained is presented in comparison with a current period of operation in Table 9.

Operating mode	**CO_2 (%)**	**CO (mg/Nm^3)**	**NO_X (mg/Nm^3)**	**SO_2 (mg/Nm^3)**	**HCl (mg/Nm^3)**	**HF (mg/Nm^3)**	**PM (mg/Nm^3)**
Test A	**17.64**	**1372.31**	474.49	38.38	0.22	0.66	0.97
Test B	**17.17**	**1003.51**	466.89	28.91	0.25	0.58	0.79
Current operation	**17.71**	**1360.59**	471.30	30.67	0.06	0.65	0.81
Limit AIM	**-**	**2000**	**500**	**400**	**10**	**1**	**30**

Table 9. Pollutant emissions at the stack of the clinker plant

All pollutant concentration values resulting from the monitoring data of the plant are within the limits of the AIM, with variations in a relatively constant range. It can be stated that the slag used as raw material does not influence the values of pollutant concentrations in emissions. As far as the measured CO_2 emissions are concerned (momentary values), a decrease of the CO_2 concentration in the exhaust gas of approx. 0.4 % during test A and approx. 3 % during test B, the introduction of slag at the cold head of the furnace having a positive effect on the reduction of CO_2 emissions and thus on the environment.

5. CONCLUSIONS

Measurements of CO_2 at the chimney of the clinker plant showed a decrease in CO_2 concentration in the exhaust gas of approx. 0.4% during test A and approx. 3 % during test B. This demonstrates that the introduction of slag at the cold head of the furnace has a positive effect on the reduction of CO_2 emissions and thus on the environment. From the point of view of the quality of the clinker obtained experimentally, the following is observed:

- slightly lower values for calcium oxide saturation (by about 1.5 p.u.) and alumina modulus (MAl: 1.33 – 1.36 vs. 1.43) compared to the clinker of the current operating period;
- higher content of dicalcium silicate (C_2S = $2CaOSiO_2$) compared to the clinker of the current production run (C_2S: 14.5 – 15.4 % compared to 11.9 %);
- lower free CaO content compared to the clinker of the current production run (1.13 – 1.35 % vs. 1.43 %);
- compliance with the conditions laid down in SR EN 197-1, namely: $C_3S + C_2S$ = 74.77 – 77.84 % compared to 2/3 of the required mass; CaO/SiO_2 = 3.01 – 3.20 compared to the minimum 2 required; MgO = 1.52 – 1.86 % compared to the maximum 5 % required (where: C_2S – dicalcium silicate $2CaOSiO_2$, C_3S – tricalcium silicate $3CaOSiO_2$).

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