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New types of low-carbon cements with reduced Portland clinker content as a result of ecological actions of cement industry towards sustainable development

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Abstract: Cement industry is among the most energy-intensive economic sectors. Moreover, significant amounts of carbon dioxide are released to the atmosphere as a result of Portland clinker combustion process, where carbonates (limestone, marl) are the main raw materials applied. To offset the harmful effects of the cement industry on the environment, cement producers have resorted to various ways to reduce greenhouse gas emissions and to reduce energy consumption required to produce cement clinker. Development trends in the area of binding materials aim therefore towards a significant reduction of the Portland clinker content in cements for mineral additives. The outcome of these efforts is an amendment plan to the cement standard EN 197-1 based on the introduction of new composite cement groups CEM II/C and CEM VI characterized by significantly reduced amount of Portland clinker (i.e. ternary cements). The essence of actions takes is to obtain composite cements of a good quality, which, in the wake of the selection of relevant non-clinker constituents combinations, will become an alternative for traditional cements of common use.

Hereby paper presents the research results of the 4 new cement types, which confirm the relevance of further development of pro-ecological composite cements.

Keywords: ternary cements, mineral additives, sustainable development, carbon dioxide emission, cement industry

JEL codes: L61, Q42

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1. Introduction

Sustainable development is moving towards efficient economy resources, greener and more competitive. This implies building a low carbon economy, with cost efficient and resource-effective manner, with the consequence, protecting the environment by reducing greenhouse gas emissions. The above-mentioned operations are carried out intensively in the cement industry. This is mainly due to the increasingly strict legislation on climate and energy policy of the EU, by which Member States are obliged to progressively reduce carbon dioxide emissions in the industry. The share of total CO_2 emissions in 2014 for industrial processes (IPPU) in Poland was 6.6%. In this category, the main source of emissions are "mineral products" - especially cement production (National Inventory Report, 2016). The cement industry is one of the biggest areas with carbon footprint, which mainly results from the technology of Portland clinker production (cement industry accounts for around 5% of global carbon dioxide (CO_2) man-made emissions).The emission of carbon dioxide in cement industry has its grounds in the two basic and direct sources: decarbonisation process of calcium carbonate and fossil fuel combustion, moreover in the two the indirect ones: production of electricity used in cement plant and transport (production of its one tone normally generates the emission of 600-700 kg CO_2) (SPC, 2015; Chłądzyński and Garbacik, 2008). To meet the demands of the EU and at the same time to maintain production of cement at a stable level, it is necessary to limit the raw material clinker in the cement composition and replace it with other components, which perform hydraulic and/or pozzolanic properties. These are the most well-known and widely used in cement and concrete production technological waste products from other industries, i.e. fly ash from power plants, granulated blast furnace slag from iron or silica fume from ferrosilicon production. Ground limestone acting also as a partial replacement for cement, is widely available and can be sourced from own cement plant pits (Chłądzyński and Garbacik, 2008; Giergiczny et al., 2009:30-35). Although, there is no shortage of cements with mineral additives on the market of binding materials, pro-environmental trends, as well as constant striving to improve them with better and better properties are conducive to their further growth and product range development. The article presents the test results of the new types of ternary cements and concretes made thereof. The aim of the study is to confirm that cements with low content of Portland clinker replaced by a mixture of a large amount of technological waste perform adequate quality and are an alternative to traditional cements with a large content of production-expensive Portland clinker.

2. The role of cement in low-carbon economy

Cement is primarily used as a binder in concrete, which in turn, is the essential material in all building types (public and residential buildings, roads, bridges, dams, etc.). Reduction of the content of clinker in cement composition results in its decreased demand in concrete production, and thus, has a direct impact on lowering the production cost of concrete of and emerging construction. Noting the energy efficiency of the building it is important to point out a significant role of concrete. Its thermal inertia means that wisely developed modern concrete buildings can consume 75% less energy throughout the lifecycle. Therefore, the method and time of use of concrete can have a huge impact on global emissions. Cement industry is a leader in research on concrete, its development, production and technology. With its innovative use of concrete products, the contribution of an industry to a low carbon economy in 2050 can go far beyond the reduction of emissions related to the production of cement (SPC, 2015; Chłądzyński and Garbacik, 2008). As a key component of concrete, cement will have to play a very important role in the economy of resources and solving the problems arising from a growing population and urbanization.

The most key investments realized with the essential participation of cement are:
- innovative buildings to provide energy-efficient home or workplace, while serving as facilities generating renewable energy,
- new transport solutions that minimize impact on the environment and reducing congestion,
- vertical buildings to reduce the surface area required for a growing population,
- major investments to help use the energy of wind, tide and sun,
- infrastructure that helps to protect people from the possible rise of sea levels (SPC, 2015).

Concrete will be the main material of choice for the application of most of these solutions.

3. Reduction strategies of CO_2 emission by cement industry

As mentioned previously, there are four main sources of CO_2 emissions in the industry, regarded as independent of each other. Therefore, CO_2 emissions in the cement industry can be reduced by (Baran et al., 2010:22-26):

- improving the production process, that is improving the performance of cement kilns; replacing energy-intensive wet methods with dry and semi-dry one;
- modernization of a cement plant in order to reduce electricity used, greater concentration of production in a more efficient plants and the recovery of heat energy generated in production processes and its application in energy production used later in technological processes.
- through the valorization of wastes in production processes or the use of waste as alternative fuels and thus, eliminating the disposal by landfill sites and waste incineration (CEMBUREAU, 2004); the use of waste as raw material in clinker production, reducing the amount of clinker per ton of cement by the usage of industrial waste, for example: granulated blast-furnace slag, fly ash, natural pozzolans as cement components. It should be noted that the use of industrial waste as mineral additives to cement can improve the quality of products, which generates growth in strength and durability, thereby improving the efficiency of cement use (e.g. increased density of concrete containing blast furnace slag results in improved resistance to the effects of corrosive environments as well as the increased strength after longer periods of setting).

All these measures are consistent with a policy of sustainable development.

4. New types of low-carbon cements CEM II/C-M and CEM VI

European Standardization Committee is working on widening the range of common cements within the scope of EN 197-1. A characteristic feature of these composite cements is a higher concentration of a mixture of the two non-clinker main components in their composition. Such binders are present in ternary combinations K-S-L/LL,K-S-V and K-V-L/LL (K- clinker, S-granulated blast furnace slag, V-siliceous fly ash, L/LL- lime). Current cement compositions will therefore be extended by an additional subgroup of CEM II/C, with increased share of mineral additives up to 50%, and by a whole new group of composite cements CEM VI, of which the content of Portland clinker would not exceed 50%. The content of the main non-clinker components would constitute within this group from 50 to 65%. Hitherto group of CEM V would then change its name from composite cements into pozzolanic – slag cements (Table 1) (pr EN 197-1: 2014; Giergiczny and Sybilski, 2014:3-5; Kuterasińska and Król, 2015:58-61).

Table 1. New types of common cements acc. to pr EN 197-1 (2014)

Cement type	Designation		Component share in cement, % by mass				
			Clinker K	Blast furnaceslag S	Natural Pozzolan P	Fly ash V	Lime L and LL
CEM II	Portland composite cement	**CEM II/C-M (S-L)**	50-64	16-44	-	-	6-20
		CEM II/C-M (S-LL)		16-44	-	-	6-20
		CEM II/C-M (P-L)		-	16-44	-	6-20
		CEM II/C-M (P-LL)		-	16-44	-	6-20
		CEM II/C-M (V-L)		-	-	16-44	6-20
		CEM II/C-M (V-LL)		-	-	16-44	6-20
		CEM II/C-M (S-V)		16-44	-	6-20	-
CEM VI	Composite cement	**CEM VI (S-L)**	35-49	31-59	-	-	6-20
		CEM VI (S-LL)		31-59	-	-	6-20
		CEM VI (S-V)		31-59	-	6-20	-

Source: (pr EN 197-1, 2014)

The expansion of composite cements assortment is justified primarily by the fact that one can form their properties using the synergy influence effect of the mineral additives applied. It is a common knowledge that the effect of individual non-clinker main components on cement properties, can vary. A simple example of such is the influence of the fly ash and ground limestone addition on strength development. Siliceous fly ash retards the development of strength in early setting times, but do not reduces the strength in longer periods. In contrast, lime accelerates the development of early strength, but reduces the strength in the longer periods of setting. With a combination of various mineral additives in composite cements, it is possible to use their favourable characteristics, furthermore, by optimizing the composition of cement mixture, their adverse impact can be effectively suppressed (Giergiczny, 2014).

5. Performance characteristics of the new ternary cement types CEM II/C-M and CEM VI

5.1. Test materials

Granulated blast furnace slag S, siliceous fly ash V and limestone LL were used as mineral additives in the tests. As the clinker carrier in cement composition, an intermediate (chemical and phase composition - Table 3) with increased content of SO_3 (5.1%) was applied. Tables 2 and 3 show a description of the chemical composition and some physical properties of the respective constituents.

Table 2. Chemical composition and physical properties of non-clinker main cement compounds

Compound	Compound content [%]									Density[g/cm³]	Blaine surface [cm²/g]
	SiO_2	AL_2O_3	Fe_2O_3	CaO	MgO	SO_3	Na_2O	K_2O	Cl^-		
Limestone (LL) *)	5,35	1,28	1,07	49,7	1,8	0,03	< 0,1	<0,1	0,005	2,70	6150
Granulated blastfurnace slag (S)	40,51	7,38	1,26	43,7	5,03	0,14	0,77	0,45	0,046	2,92	3795
Siliceous fly ash(V)	52,33	27,48	5,80	3,58	2,61	0,29	0,94	3,15	0,008	2,14	2761

*) $CaCO_3$content calculated onCaO base content and amounts 89%, Total organic carbon (TOC)- 0,04%, clay content-0,4g/100g

Source: Author's own elaboration..

Table 3. Chemical composition and phase intermediate (designated in the text as C)

Chemical composition [%]										Phase composition [%]			
Loss on ignition	SiO_2	AL_2O_3	Fe_2O_3	CaO	MgO	SO_3	Cl^-	Na_2O	K_2O	C_3S	C_2S	C_3A	C_4AF
1,54	20,6	5,09	2,57	62,94	1,38	5,10	0,09	0,15	0,63	49,9	28,5	10,3	8,8

*) intermediateacc. to Blaine surface 6200 cm²/g

Source: Author's own elaboration..

Mineral additives applied in the study conform to the requirements of PN-EN 197-1: 2012. Ternary cements were obtained by mixing the components with an intermediate in adequate proportions (homogenization in a laboratory mixer). For the purpose of comparison, cement with the one non-clinker component was performed to show the synergistic effect of additive mixture on the properties of ternary cements. The composition of tested cements is presented in Table 4. Table 5 illustrates the content of selected chemical compounds of ternary cements.

Table 4. Composition of analysed cements

Cement designation	Cement type	Compound content [% by mass]			
		C	S	LL	V
I	**CEM II/C-M (30%S-10%LL)**	60	30	10	-
II	**CEM II/C-M (30%V-10%LL)**	60	-	10	30
III	**CEM VI (35%S-20%LL)**	45	35	20	-
IV	**CEM *VI* (35%S-20%V)**	45	35	-	20
CEMS*(55%S)		45	55	-	-
CEM V* (40%V)		60	-	-	40
CEM LL*(40%LL)		60	-	40	-

*) comparative cements

Source: (pr EN 197-1, 2014)

Table 5. Selected chemical properties of ternary cements CEM II/C-M and CEM VI

Cement designation	Cement type	Compound content [%]						
		L.O.I.	Insoluble residue	Cl^-	SO_3	Na_2O	K_2O	Na_2O_{eq}
I	**CEM II/C-M (30%S-10%LL)**	5,62	0,53	0,10	3,06	0,28	0,56	0,65
II	**CEM II/C-M (30%V-10%LL)**	6,43	22,52	0,09	3,21	0,39	1,36	1,28
III	**CEM VI (35%S-20%LL)**	8,98	0,98	0,09	2,26	0,31	0,48	0,63
IV	**CEM *VI* (35%S-20%V)**	1,14	11,4	0,08	2,41	0,49	0,97	1,13

Source: Author's own elaboration..

5.2 Test methods

The scope of research and designations covered cement properties required by the standard PN-EN 197-1 (2012). Physical and mechanical properties of cement according to the procedures contained in the PN-EN 196 (2006; 2011), i.e. water demand, setting time, soundness (LeChatelier) and compressive strength were analyzed. The consistency of standard mortars were determined by the slump test method in accordance with PN-EN 1015-3 (2000). Concrete tests in terms of compressive strength and frost resistance F150, in line with the procedures given by standards in PN-EN 12390-3 (2011) and PN-88/B-06250 (1999), were also performed.

5.3. Test results and discussion

5.3.1. Cement properties

The test results of cements physical properties are listed in tables6and 7 and illustrated in Figure 1-2.

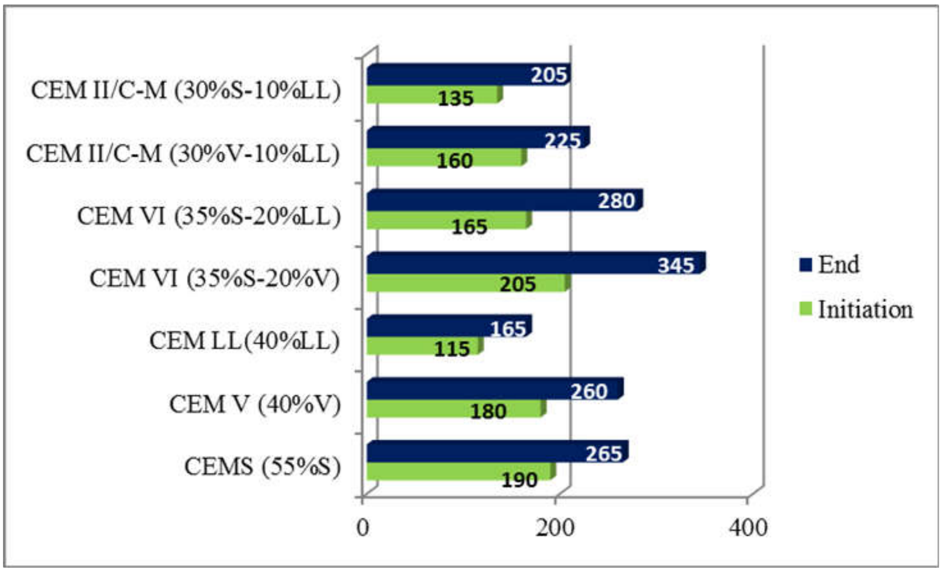
Table 6. Physical properties of analysed ternary cements.

Cement designation	Cement type	Proper water amount	Setting time, [min]		LeCh	Density	Blaine surface
		[%]	Beginning	End	[mm]	$[g/cm^3]$	$[cm^2g]$
I	**CEM II/C-M (30%S-10%LL)**	32	135	205	0,5	3,00	4730
II	**CEM II/C-M (30%V-10%LL)**	29	160	225	0	2,71	4710
III	**CEM VI (35%S-20%LL)**	32	165	280	0	2,96	4631
IV	**CEM VI (35%S-20%V)**	32	205	345	0	2,82	4363
CEMS*(55%S)		33	190	265	0	2,98	4330
CEM V* (40%V)		29	180	260	0	2,64	4300
CEM LL*(40%LL)		30	115	165	1	2,92	5765

Source: Author's own elaboration..

The longest period of the beginning of setting time is specific to composite cement CEM VI (S-V) (IV) containing 20% of fly ash and 35% of granulated blast furnace slag. It is close to the beginning of CEM S cement with 55% of granulated blast furnace slag (typical content for blast furnace cement CEM III/A) and CEM V, containing 40% of siliceous fly ash addition. The combination of slag and fly ash in cement composition increases the setting time with respect to cement containing only one of these additives. It can be observed that the shorter setting times (Table 6 and Figure 1) are characteristic for cements with a higher content of cement clinker and ground limestone. A typical feature of these materials is also higher specific surface area, being the result of the presence of ground limestone in cement composition (Table 2).

Figure 1. Setting times of ternary cements CEMII/C-M and CEM VI in relation to cements containing one mineral additive.



Source: Author's own elaboration.

The lowest water demand (29%) was performed by Portland-composite cement CEM II/C-M (V-LL) and cement with siliceous fly ash addition CEM V (Table 6). Low water demand of Portland fly ash – lime cements arises from the synergistic effect of ground limestone (inert additive) and fly ash, characterized by a high content of glassy phase and spherical shape of grains (Kuterasińska and Król, 2015:58-61). Ternary cements consisting of slag and ground limestone are characterized by higher water demand. This is due to the relatively high activity of granulated blast furnace slag and high specific surface area of ground limestone. However, in modern concrete technology, increased water demand of concrete mix arising from, among others, the water demand of cement, is effectively reduced by the use of admixtures lowering the amount of water in concrete mix (plasticizers and superplasticizers).

Therefore, it may be assumed that the ternary cements have a consistency similar to one of mortars produced from cements with the addition of a one mineral additive (Table 7).

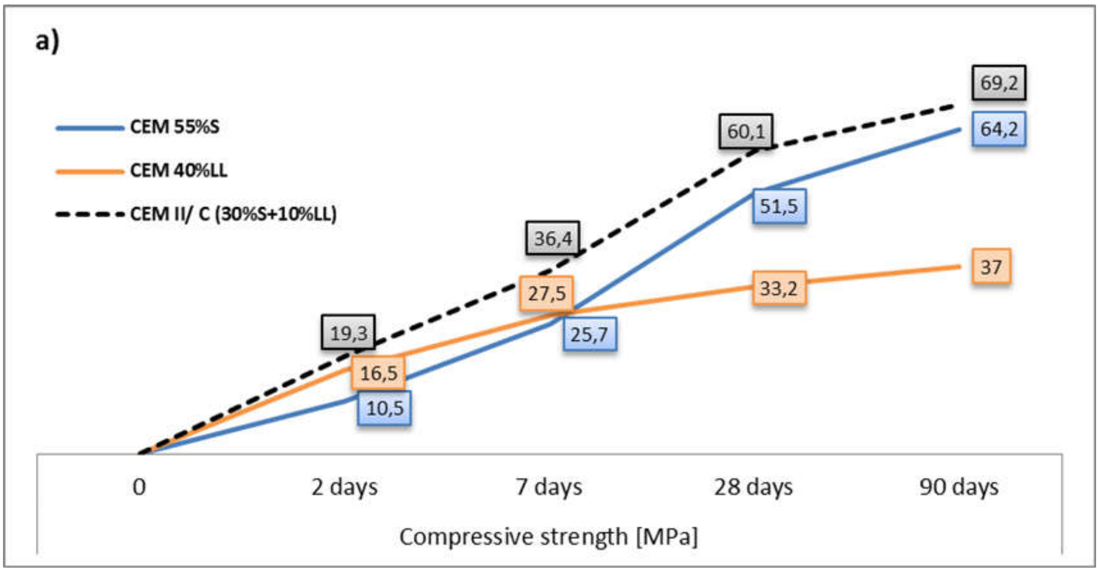
Table 7. Mechanical properties and consistency of mortars made from CEM II/C-M and CEM VI and cements containing one mineral additive

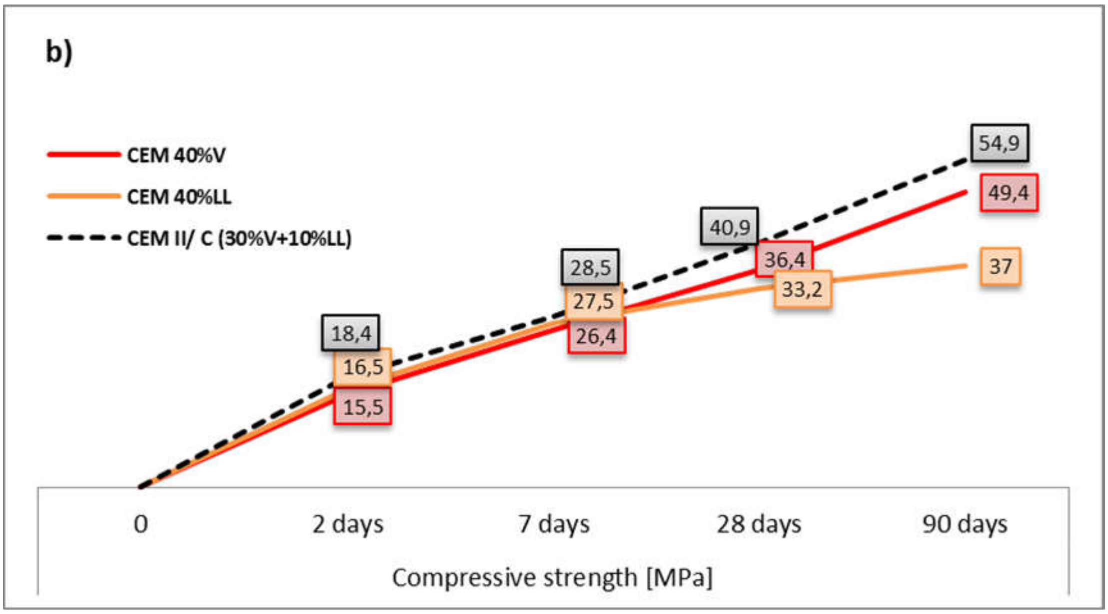
Cement designation	Cement type	Consistency [mm]	Bending tensile strength [MPa]				Compressive strength [MPa]			
			2 days	7 days	28 days	90 days	2 days	7 days	28 days	90 days
I	CEM II/C-M (30%S-10%LL)	155	4,0	6,7	8,9	9,7	19,3	36,4	60,1	69,2
II	CEM II/C-M (30%V-10%LL)	163	3,7	5,2	6,6	8,5	18,4	28,5	40,9	54,9
III	CEM VI (35%S-20%LL)	158	2,8	5,5	8,2	9,3	10,6	25,0	46,8	56,2
IV	CEM VI (35%S-20%V)	156	2,3	5,0	7,1	9,4	10,7	22,8	45,4	59,1
CEM S		152	2,4	5,7	8,7	10,0	10,5	25,7	51,5	64,2
CEM V		162	3,1	4,5	6,0	6,9	15,5	26,4	36,4	49,4
CEM LL		155	3,3	5,0	5,9	6,2	16,5	27,5	33,2	37,0

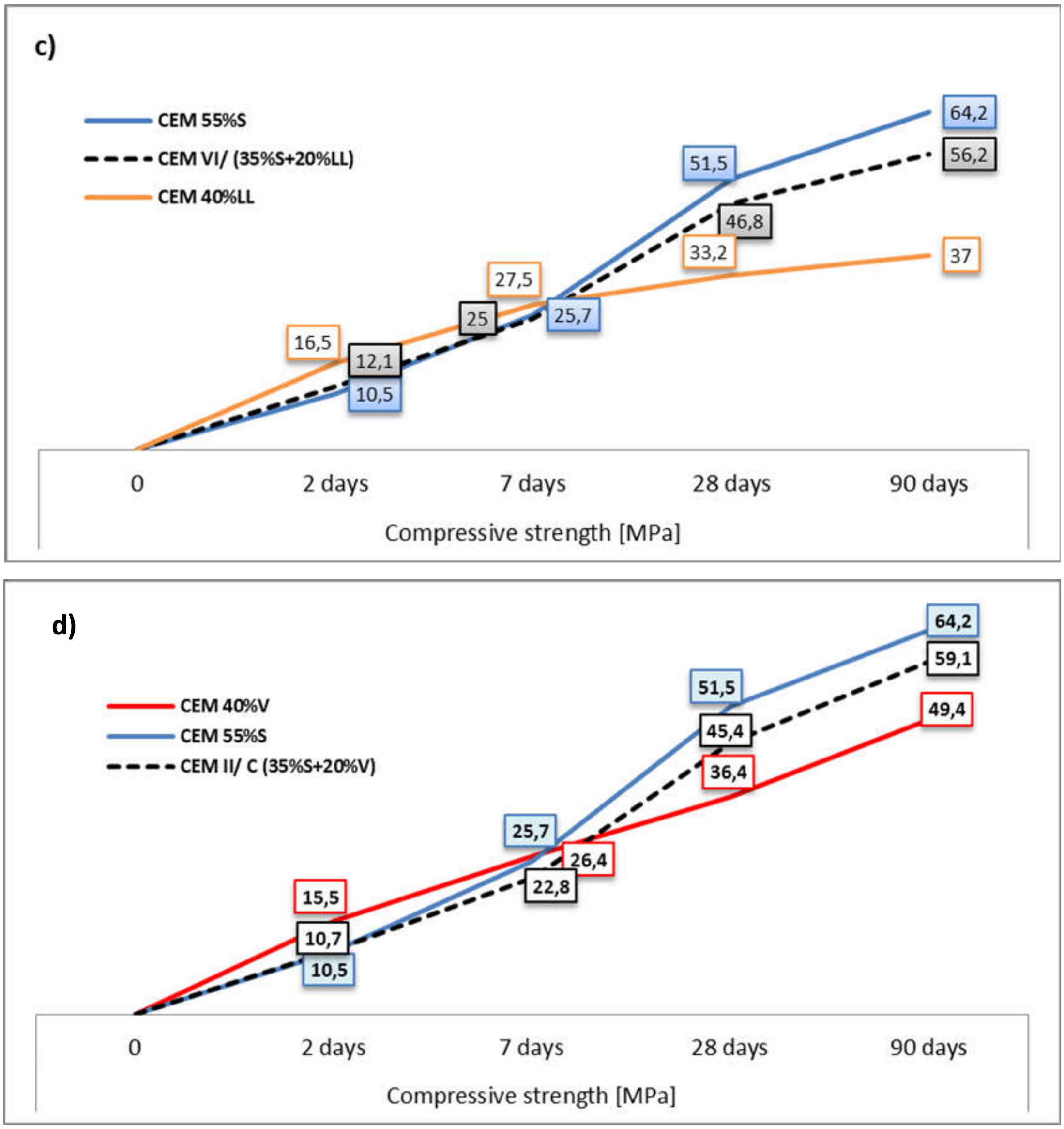
Source: Author's own elaboration.

The analysis of strength properties of tested cements allow to state that ternary cements CEM II/C (S-LL) and CEM VI (S, LL), containing ground granulated blast furnace slag and ground limestone have the highest strengths, and can be classified as strength class 42.5 N according to PN-EN 197-1 [SPC, 2015], whereas Portland composite cement CEM II/C-M (V-LL) meets the requirements of strength class 32,5 R. Portland slag – lime cement CEM II/C-M with 40% content of additives (30%S + 10%LL) performed the greatest compressive strength, in all studied periods (Figure 2a). Standard strength (after 28 days) and long term strength (after 90 days) of composite cements: slag – lime cement CEM VI (35% S –20%LL) and slag – fly ash cement CEM VI (35% S -20% V) with a lower content of clinker (55% of mineral additives) significantly exceeds the strength of fly ash- lime cement CEM II/C-M (30% V-10% LL) - table 7, containing more clinker in its composition. Comparing the compressive strength of cements with one type of mineral additive (CEM V, CEM S and CEM LL) with the strengths of ternary cement CEM II/C-M and CEM VI, the effect of synergistic influence of the mixture of mineral additives used can be observed (Figure 2).

Figure 2. Compressive strength of cement [MPa]: a) CEM II/C-M (30% S, 10% LL); b) CEM II/C-M (30% V, 10% LL); c) CEM VI (35% S, 20% LL); d) CEM VI (35% S, 20% V) in comparison with one mineral additive cement (CEM 55%S, CEM 40%V, CEM 40%LL)







Source: Author's own elaboration.

Fly ash–lime ternary cement CEM II/C-M (30%V-10%LL) enhanced better mechanical properties than fly ash cement and calcareous cement (Figure 2b). This may result from the sealant effect of ground limestone, which has a much higher specific surface area (Kuterasińska and Król, 2015).

Portland cement CEM II/C-M and composite cement CEM VI involving ground granulated blast furnace slag and ground limestone are characterized by significantly improved strengths than calcareous cement (CEM LL) with alike characteristics of strength as compared to slag cement CEM S with a slag content of 55% (Figure 2a and 2c). This is due the combined properties of ground limestone (sealant effect) and high-activity of ground granulated blast furnace slag. The synergy of mineral additives interactions is also visible in slag – fly ash cement (Figure 2d).

5.3.2. Concrete properties

To check the feasibility of new types of ternary cement practically, concrete containing these cements was prepared. The mixture was made in two variants: with a water/cement ratio equal to w/c = 0.6 and w/c = 0.35, wherein polycarboxylate ether superplasticizer was applied. Selected compositions of cement were chosen in order to obtain constant percentage contain of individual mineral additive, whereas only their types might vary. This will allow to look at the synergy effect between various components. The table below shows the composition of the concrete.

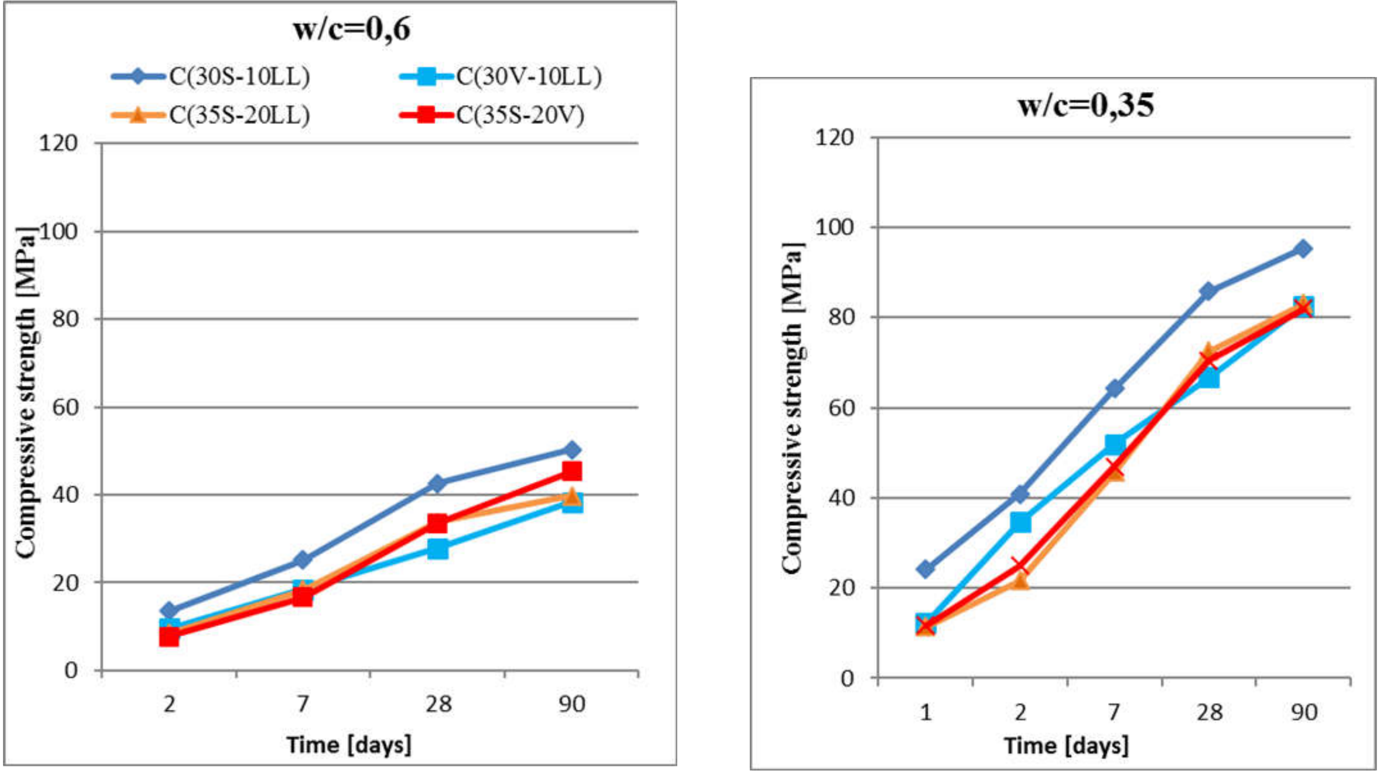
Table 8. Concrete composition

Component	**Content, kg/m^3**	
	w/c=0,6	**w/c=0,35**
Cement	300	340
Water	180	120
Sand 0-2	680	725
Gravel 2-8	530	565
Gravel 8-16	680	725
Polycarboxylate ether superplasticizer	-	10,2

Source: Author's own elaboration.

Figures 5a and 5b illustrate the compressive strength of concretes.

Figure 3. Compressive strength of concretes a) w/c=0,6 and b) w/c=0,35 (with superplasticizer)



Source: Author’s own elaboration.

The best strength parameters achieved concrete with slag – lime cement with 40% content of mineral additives. Replacement of slag with siliceous fly ash (V) in combination with limestone (LL) results in deterioration of concrete strength (decrease of approximately 25%). In case of concrete with 55% content of mineral additives the development of early strength and after 28 days looks very similar, but after 90 days of setting, the strength of concrete with slag – lime cement decreases by approx. 15%. The reduction of mixing water by the application of waterproofing admixture demonstrates how favorable results can be achieved by using concrete admixtures. Thus, obtained concrete strength indicate good compatibility of used ternary cements with plasticizers and give the possibility to produce high strength concrete with their addition.

6. Conclusion

In the last decade, the impact of cement plants on the environment has been dramatically reduced by thorough technical modernization. Emission of dust, which is most commonly associated with the image of a cement plant, has become insignificant. Eliminating the wet method of cement clinker production led to a reductions thermal energy use, and thus, the amount of emitted gases. This made it possible to greatly reduce CO_2 emissions about 40% by combusted clinker unit compared to the early nineties. The activities of cement industry also bring environmental benefits. Cement production consumes about 4 million tonnes of waste per year, so that waste is eliminated from the environment, hence, deposits of raw materials and non-renewable fossil fuels are preserved. With regard to the research on new types of cements with high content of non-clinker additives it can be concluded that these are the binders of a good quality. Therefore, the desirability of developing new assortment of composite cements CEM II/C-M and CEM VI types has been confirmed. The properties of these cements and concretes achieved with their addition are both the outcome of component properties and compositions thereof (S-LL, V-LL, V-S) as well as their mutual proportions. The application of the maximum amount of mineral additives allows obtain ternary cements in classes 32.5 R and 42.5 N. Proposed configuration of the three components in CEM II/C-M and CEM VI compositions provides better or comparable mechanical and physical properties in comparison with the properties of cements with one mineral additive, due to the synergy effects of these additives interactions. The concretes obtained with $w/c = 0.35$ are characterized by very high strengths, qualifying them to a group of high strength concretes. Integrated operations of cement industry aiming at the reduction of clinker content in cement composition in favour of technological waste, moreover, both the improvement of clinker production technology and the application of alternative fuels in its production prove sustainable development policy in the cement industry.

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Nowe rodzaje niskoemisyjnych cementów o zredukowanej zawartości klinkieru portlandzkiego jako rezultat proekologicznych działań przemysłu cementowego na rzecz zrównoważonego rozwoju

Streszczenie

Przemysł cementowy jest zaliczany do jednych z najbardziej energochłonnych sektorów gospodarki. Co więcej, w wyniku wypalania klinkieru portlandzkiego w cementowniach, gdzie głównymi zastosowanymi surowcami są węglany (kamień wapienny, margiel), do atmosfery uwalniane są ogromne ilości dwutlenku węgla. Aby zniwelować szkodliwe oddziaływanie przemysłu cementowego na środowisko naturalne, producenci cementu uciekają się do różnych sposobów ograniczenia emisji gazów cieplarnianych oraz do ograniczenia zużycia energii niezbędnej do produkcji klinkieru cementowego. Trendy rozwojowe w zakresie spoiw wiążących zmierzają więc w kierunku znacznej redukcji zawartości klinkieru portlandzkiego w cementach na rzecz dodatków mineralnych. Rezultatem tych starań jest plan nowelizacji normy cementowej EN 197-1 polegający na wprowadzeniu nowych grup cementów wieloskładnikowych CEM II/C i CEM VI charakteryzujących się znacznie zredukowaną zawartością klinkieru portlandzkiego (tzw. cementy trójskładnikowe). Istotą podjętych prac jest uzyskanie cementów wieloskładnikowych o dobrej jakości, które w wyniku doboru odpowiednich kombinacji składników nieklinkierowych, będą stanowiły alternatywę dla tradycyjnych cementów w powszechnym zastosowaniu. W prezentowanej pracy przedstawiono wyniki badań dla 4 rodzajów cementów nowego typu, które potwierdzają słuszność dalszego rozwoju ekologicznych cementów wieloskładnikowych.

Słowa kluczowe: cement trójskładnikowe, dodatki mineralne, zrównoważony rozwój, emisja dwutlenku węgla, przemysł cementowy