

AN OVERVIEW OF INDUSTRIAL WASTES AS FUEL AND MINERALIZER IN THE CEMENT INDUSTRY

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Abstract— The cement industry worldwide is facing growing challenges in conserving material and energy resources. According to the International Energy Agency, the main levers for cement producers are the increase in energy efficiency and the use of alternative materials. This work shows the application of industrial wastes in cement industry as fuel and mineralizer that are found out in many studies of literature. The works showed that the waste fuels are viable but it is necessary to take to account that the burning behavior of most waste fuels differs significantly from the behavior of fossil fuels due to higher particle sizes, material densities and transport characteristics. On the other hand the mineralizer decreases of burning temperature of Portland clinker without altering the clinker quality.

Keywords— industry waste, clinker, mineralizer

I. INTRODUCTION

The cement production has undergone development from 2000 year. The annual global cement production has reached 2.8 billion tons and is expected to increase to some 4 billion tons per year (Schneider *et al.*, 2011). The typical electrical energy consumption of a modern cement plant is about 110–120kWh per ton of cement (Rahman *et al.*, 2013).

Cement is one of the most important building materials worldwide, it is used mainly for the production of concrete, then the cement consumption and production is closely related to construction activity and, therefore to economic activity.

Cement sub-sector represent an energy intensive industry, approximately 12 – 15% of total industrial energy use. The typical primary energy consumption for cement production is 75% fuel and 25% electrical energy (Madloul *et al.*, 2011). In the manufacturing process thermal energy is used mainly during the burning process, while maximum share of electrical energy is used for cement grinding (Rahman *et al.*, 2013).

Forward this high fuel consumption the cement companies are introducing the industrial waste co-processing from a perspective that combines environmental policies with the interests of companies. The industrial wastes can provide part of the energy required to make cement.

The production of energy from alternative sources and its impact on climate change are among the main strategic in the sustainable development. Various types of wastes are used towards the production of energy and

the reduction in the use of fossil fuels (Samolada and Zabaniotou, 2014).

The use of wastes in the cement industry has another advantage, which is the property of mineralizer some wastes. This incorporation in small proportions improves the clinkering conditions as well as decreasing the maximum clinkering temperature, or improves the phase formation in the clinker without altering the final properties of the product (Giménez-Molina *et al.*, 1995).

In this context the present different work present a literature review of cement production applying industrial wastes as fuels and mineralizers.

First a review of cement production was developed, after it was searched the main wastes fuel used by industries and their quantities applied and their effects in clinker produced.

Finally the work concentrated to show the advantages of using the mineralizers, as example, they are responsible by decreasing the thermal energy consumption.

II. PORTLAND CEMENT PRODUCTION

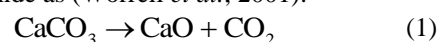
Portland cement is made by heating a mixture of limestone and clay, or other materials of similar bulk composition and sufficient reactivity (Taylor, 1964).

The raw materials must be crushed and proportioned so that the appropriate chemical composition of the raw material feed is reached (Lamas *et al.*, 2013). The chemical composition and the size distribution are crucial to both the quality of the product and the operation of the process (Kosmatka *et al.*, 2002).

After crushing the raw material is sent to pyro processing stage (to a temperature of about 1450°C). Partial fusion occurs, and nodules of clinker are produced (Taylor, 1964).

There is a combination of endothermic and exothermic reactions occurring in an extremely complicated chemical reaction sequence. In essence the following material transformation occurs in pyro processing stage (Table 1).

From Table 1 is described the raw material transformation, one important phase of the production of the clinker is the calcination process. In this chemical reaction calcium carbonate decomposes in calcium oxide and carbon dioxide as (Worrell *et al.*, 2001):

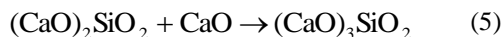
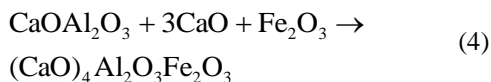
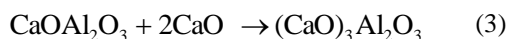


The main compound of cement clinker is the Calcium oxide (CaO), this compound (inside the kiln) reacts with other oxides such as: silica, alumina, iron oxide for the formation of $3\text{CaO} \cdot \text{SiO}_2$ (C_3S), $2\text{CaO} \cdot \text{SiO}_2$

Table 1 – Material transformation in pyro processing stage (Bhatty *et al.*, 2004).

Process	Temperature
1. Evaporating free water, at temperature up to	100°C
2. Removal of adsorbed water in clay materials	100 – 300°C
3. Removal of chemically bound water	450 – 900°C
4. Calcination of carbonate materials	700– 850°C
5. Formation of C ₂ S, aluminates and ferrites	800 – 1250°C
6. Formation of liquid phase melt	>1250°C
7. Formation of C ₃ S	1330 – 1450°C
8. Cooling of clinker to solidify liquid phase	1300 – 1240°C
9. Final clinker microstructure frozen in clinker	< 1200°C
10. Clinker cooled in cooler	1250– 100°C

(C₂S), 3CaO.Al₂O₃ (C₃A) and 4CaO.Al₂O₃.Fe₂O₃ (C₄AF). The main chemical reactions are presented below (Çamdali *et al.*, 2004):



The C₃S and C₃A are responsible for the strength of the cement. The high percentages of C₃S result in high early strength but also high heat generation as the concrete sets. C₃A causes undesirable heat and rapid reaction properties, which can be prevented by adding gypsum, CaSO₄.2H₂O to the final product (Paula, 2009).

The kiln charge must spend sufficient time at high enough temperature to reach near complete reaction to form clinker minerals. The material bed must tumble adequately to ensure uniform heating through the mass. A summary of a typical kiln heat balance during clinkering reactions is presented in Table 2.

Table 2 – Summarized data on a typical kiln heat balance for dry process (Bhatty *et al.*, 2004)

Endothermic reactions	kJ/kg
Dehydration of clays	170
Calcination	1990
Heat of melting	105
Heating raw mats (1450°C)	2050
Sub total	4315
Exothermic reactions	
Crystallization dehydrated clay	40
Heat of formation, clinker minerals	420
Crystallization of melt	105
Cooling of clinker	1400
Cooling of CO ₂	500
Cooling of water	85
Sub total	2550
Net heat for clinker formation	1765
Heat losses related to inefficiencies during the kiln operation	
Evaporation of water	20
Heat losses, gas, clinker, dust	840
Radiation, convection loss	650
Total heat consumption	3275

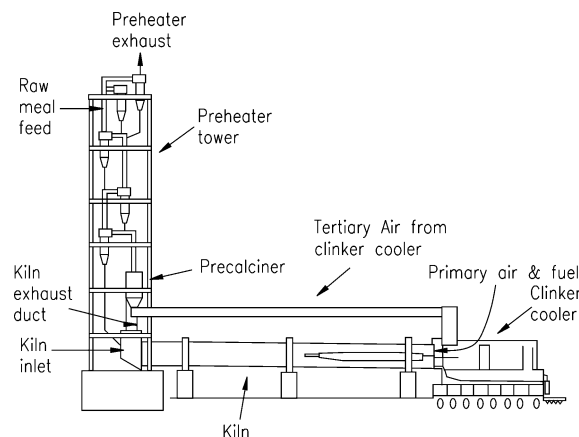


Figure 1 - Precalciner kiln of cement industry

The final step in Portland cement manufacturing is grinding the blend of clinker and gypsum. Up to 5% (by weight) of gypsum is added to the clinker after it is cooled. The amount of gypsum is adjusted to regulate cement properties such as setting time (Kosmatka *et al.*, 2002).

Depending on how the raw material is handled before being fed to the kiln, basically three different types of process can be distinguished: the dry, semi-dry/semi-wet and wet process. In dry process the feed material enters the kiln in a dry, powdered form. The dry feed is preheated in a tower of heat exchange cyclones in which the dry feed is preheated by hot exit gases from rotary kiln prior to entering the kiln.

The calcination process can almost be completed before the raw material enters the kiln if part of the fuel is added in a special combustion chamber (precalciner kiln) as it showed in Fig. 1 (CEMBUREAU, 1997).

In the wet process the feed material goes to wet grinding and the resulting slurry, which contains typically 30 – 40% water. In the semi-dry or semi-wet process 10 – 20% water is either added to the ground dry feed material resulting in a feed material containing about 15 -20% moisture (CEMBUREAU, 1997). Over past few decades the worldwide cement industry has invested in moving from wet process to the more energy-efficient dry process

III. WASTE FUEL IN CEMENT INDUSTRY

Nowadays the high degree of the industrial activity as well as the increasing standards of living has been accompanied by a growing waste generation which represents a serious environmental problem. The possibility to use some industrial wastes in the cement production as source of raw materials or alternative fuel have been viable to cement industries production cost.

Cement kilns and calciner have technical conditions very favorable for use of alternative fuels, as example, the temperatures in the kilns of approximately 1500°C are higher than the threshold ignition temperature, as established by environmental regulations, long retention time of products under high temperature combustion, the high alkalinity atmosphere readily absorbs most acidic gases released by the oxidation of sulphur and chlorides (Greco *et al.*, 2013).

Table 3 – Application of waste fuel in cement industry (Rahman *et al.*, 2013).

Company	% of Thermal energy from waste fuel
Holcim Group	12.2
Cemex Group	24.7
Heidelberg Group	21.2
Italcementi Group	5.8
Lafarge Group	13

The waste fuel utilization in cement kiln is still processing, the advantage of cement kiln is the high temperatures reached inside (about 1450°C) combined with the highly oxidizing environment and the large residence time of material exposed to these conditions, represent an alternative recognized form of thermal destruction of industrial wastes (Mehta, 1979).

While in some kilns up to 100% replaced rates have been achieved, in others process places, local waste markets and permitting conditions do not allow for higher rates of waste fuel. Table 3 summarized the percentage of waste fuel applied in fuel mix by some cement industries in year 2011.

The waste fuel utilization requires the adaptation of the combustion process. Modern multi-channel burners designed for the use of alternative fuels and thermograph systems allow control of the flame shape to optimize the burning behavior of the fuels and the burning conditions for the clinker (Wirthwein and Emberger, 2010).

The wastes to be employed as fuels must meet a number of parameters which are required by the process itself. The alternative fuels should show the acceptable level of the following performance properties (Wzorek, 2012):

- **Performance as fuels**, including their calorific values which make the decisive parameters for the amounts of the conventional fuel to be substituted.
- **Chemical composition**, since the ash produced in the combustion process will be absorbed by the clinker product.
- **Physical properties**, which define stability of such fuels in the transport, storage and kiln feeding operations.

In a conventional preheater kiln (without precalciner), it is only possible to burn fuels in the kiln inlet with substitution rates of up to 25 to 30% (70 to 75% has to be fired in the main firing). On the other hand, in precalciner kilns usually up to 65% of the total fuel energy input is burned into the calciner. As consequence, in precalciner kilns, fluctuations in fuel quality have less impact on the kiln performance and clinker quality (Schneider *et al.*, 2011).

In relation to clinker quality, the burning behavior of most waste fuels differs significantly from the behavior of fossil fuels due to higher particle sizes, material densities and transport characteristics. Consequently it can change the temperature profile of the kiln including the clinkerization temperature, the length of the sintering zone and the cooling conditions. As result these changes

can affect the quality of the clinker, as example, burning grade of the clinker, the porosity of the granules, the crystal size of the clinker phases or their reactivity (Schneider *et al.*, 2011).

Other problem from the use of different fuels in the clinker burning is the formation of rings in the kiln caused by salts – such as KCl, K₂SO₄, Na₂SO₄, Ca₂SO₄ that are formed by the volatiles. This is also known as the cyclic phenomena, and the amounts of volatiles and especially sulfur, contained in the fuels have a direct effect on these phenomena (Pipilikaki *et al.*, 2005).

Chlorine content is one of the key concerns with waste derived fuels, since it may cause corrosion in the system due to vaporization and condensation of alkali chlorides (Reza *et al.*, 2013). As well as, dioxins have potential to form if chlorine is present in the input fuel or raw materials. Formation can be repressed, however, by the high temperatures and long residence times that are standard in cement kilns (Karstensen, 2008).

Take to account the problems mentioned the industries GTZ and Holcim Group Support Ltd provide a summary of the key considerations for co-incineration project (Murray and Price, 2011).

Waste hierarchy of co-processing:

- Waste should be used in cement kilns if there are not more ecologically and economically better ways of recovery.
- Co-processing should be considered an integrated part of waste management.
- Co-processing should be according with international environmental agreements.

Additional emissions and negative impacts on human health must be avoided:

- Negative effects of pollution must be prevented.
- Air emissions from cement kilns that burns alternative fuels cannot be statistically higher than those of cement kilns burning traditional fuels.

The quality of the cement must remain unchanged:

- The product must not be used as a sink for heavy metals.

Companies that co-process must be qualified

- Have good environmental and safety compliance records.

Implementation of co-processing must consider national circumstances

- Country specific requirements must reflect in regulations.

With the considerations the wastes usually co-processed in cement industry are (Lamas *et al.*, 2013):

- Used oil, including lubricant
- Soil contaminated with oil (fuel or lubricant), oily waste (rags, wood chips, sawdust, vegetation impregnated with oil and water, paper towel with oil)
- Solid waste compounds of non-toxic metal
- Slag molten of aluminum, iron, steel, and zinc
- Waste of non-metallic minerals
- Waste of paper, cardboard, polymerized plastic and textile materials

- Waste of acid washing of benzene, originating from distillation of coal tar coke
- Wastes from incineration or thermal treatment of contaminated soil
- Dirt solvents, asphalt emulsion (in solvent), mix resin with solvent.

The Table 4 summarizes the percentage of different wastes which are currently being used as waste fuel by a cement industry in year 2011.

One waste fuel used for many cement industries is the used tire, especially since the accumulation of used tires is an environmental liability, since the improper disposal of used tires can turn them into sources of environmental and public health problems (Caponero *et al.*, 2003). In Brazil, the National Environment Council – CONAMA approved a resolution, with the force of law, the resolution n° 258 of August 26, 1999, to regulate the final destination of waste tires. The resolution established a timetable for the destruction of these tires since 2002 (Mehta, 1979). Therefore the use of scrap tires as waste fuel is considered, among the ways to recycle tires, as the most viable means for its disposal. Actually in Brazil 57% of scrap tires that are discarded each year were intended to cement kilns (Lamas *et al.*, 2013).

But the use of tire as a fuel is limited to a maximum of 30% for both domestic and international cement industry due to the presence of heavy metals in their composition, mainly zinc. This has the effect of lowering the initial resistance, but ensures higher final strength of the cement. In study realized by (Pipilikaki *et al.*, 2005) the total tire used as fuel was 6%, it was concluded that no apparent problems occurred from the use of this waste fuel in the clinker burning.

The waste solvent and spent oil from different industries generally have high calorific value and those can be used in cement kiln. Vos *et al.* (2007) concluded that the maximum and minimum calorific values of solvent and spent oil were 29 MJ/kg and 36 MJ/kg respectively. Solvent and waste oil contain less minerals compared to petroleum cokes and coal hence a little additional raw meal is needed to ensure the quality of the cement. On the other side Vos *et al.* (2007) reported that CO₂ emission was reduced while solvents and paint sludge were applied as waste fuel. Seyler *et al.* (2005) showed a reduction in heavy metal emissions when waste solvents were mixed with fossil fuel.

The plastic solid waste is the global environmental problem, Siddique *et al.* (2008) reported that approximately 8% of plastic solid waste is incinerated and 7% is recycled leaving the 85% remainder to be landfilled. The application of plastic solid waste as fuel was studied by Swart (2007) through two scenarios. The baseline burn consisted of only coal, whereas the trial burn consisted of a blend of coal, plastics and waste tires. The results of the trial were that concrete compressive strengths increased slightly, CO emissions were reduced, and NO_x, SO₂ and VOC were increased compared to the coal only burn.

Table 4 – Percentage of different type of wastes used as fuel (Rahman *et al.*, 2013).

Waste type (%)	Holcim Group
Waste oil	5
Solvent	11
Tires	10
Impregnated sawdust	6
Plastic	9
Agricultural waste	9
Wood chip	15
Sewage sludge	2

The application of biomass fuels in cement industries had increased, especially since biomass fuels have lower concentrations of nitrogen and sulfur, oxides produced during the combustion process are significantly less than emissions produced by fossil fuels. The use of biomass in cement manufacturing is less common in industrialized countries and appears to be concentrated in more rural developing regions such as India, Thailand, and Malaysia.

Typical biomass fuels include wood, rice hulls, coffee grounds, others. One biomass waste important is the woodchips which are solid fuels made from woody biomass and are a byproduct of the timber industry. Woodchips are typically made from waste wood, sapling and logging operations (Thompson, 2012).

Other biomass fuel is the switchgrass, it is a warm-season grass and is one of the dominant species of the central North American tall prairie grass. In the United States, switchgrass is considered the most valuable native grass for biomass production. Switchgrass requires little fertilization and herbicide, the grass is tough and it has high productivity (Sami *et al.*, 2001).

The substitution rate of biomass for fossil fuel is quite feasible to 20% in cement kiln. Biomass is highly variable which makes flame stability and temperature control in the kiln difficult when it is used in higher proportions (Demirbas, 2003).

IV. EFFECTS OF WASTE FUELS

The burning behavior of most waste fuels differs significantly from the behavior of fossil fuels due to higher particle sizes, material densities and transport characteristics. This can change the temperature profile of the kiln including the sintering temperature, the length of the sintering zone and the cooling conditions.

All of these changes can affect different clinker characteristics like the burning grade of the clinker, the porosity of the granules, the crystal size of the clinker phases or their reactivity.

The amount and composition of the ashes introduced by waste fuel has unusual components into the kiln, as example is phosphorous. Phosphorous oxide (P₂O₅) is mainly incorporated in crystals of a solid solution of belite (C₂S) and C₃P (Ca₃P₂O₈). Belite with a certain amount of C₃P-component does not react with free lime at sintering conditions in the kiln, causing clusters of belite and free lime (Schneider *et al.*, 2011).

Kolovos *et al.* (2005) analyzed to introduce ZnO and CuO in a Portland cement raw mix and to study their ef-

fect on cement hydration and properties. The results show that the addition of 1.0 wt % CuO and ZnO in the cement raw mix exerts a slight retarding effect on the hydration process, especially at the early ages. This addition favored the cement strength development and does not affect the physical properties of the cements.

Trezza and Scian (2005) evaluates the physicochemical characteristics of the clinkers obtained after incorporating three different wastes in different proportions: two of them with energetic capacity—trade marked waste fuel and waste carbon of petroleum—and the third that would be confined—pyrolytic carbon. The results show a greater crystallinity of the principal clinker phases and cause the greater energy requirement of the milling. The better crystallized C_3S presence determines the better values of compression strength at early ages.

V. MINERALIZER

Mineralizers are inorganic compounds which accelerate the reactions of process in solid phase, liquid phase and solid-liquid interface. Many researches had studied the effect of different mineralizer on the decrease of burning temperature of Portland clinker. The fluorides, as example, according (Giménez-Molina *et al.*, 1995) have a favorable action on the formation of clinker minerals by decreasing the thermal energy consumption.

Examples of mineralizers studied are the pair CaF_2 and $CaSO_4$. The CaF_2 acts in the consecutive reactions of the clinkering process, reducing the $CaCO_3$ thermal decomposition temperature, altering the phases in thermodynamic equilibrium, varying the properties and temperature of melt formation (Johansen and Christensen, 1979)

The $CaSO_4$ reduces melt viscosity and consequently increases ion mobility. When the $CaSO_4$ increases in the raw mix a decrease on alite rate formation is observed. This decrease can be attributed to the decrease in the driving force for this reaction as well as to the effect of $CaSO_4$ on the C_2S stability range, which widens at the expense of the C_3S stability range (Johansen and Christensen, 1979).

The $CaSO_4$ and CaF_2 can in turn be used to partially replace the traditional fluxes Al_2O_3 and Fe_2O_3 to produce clinker with low aluminate contents at temperatures between 1300 and 1400°C.

The use of this mineralizing pair was studied in Blanco-Varela *et al.* (1995) that modeled the burnability of white cement raw mixes made using CaF_2 and $CaSO_4$ as fluxes/mineralizers. The main conclusions of this work were:

- The factor affecting the final free CaO in the clinker to the greatest extent is limestone particle size in the raw mix, followed by fluorite and gypsum content.
- The good reactivity and burnability of mineralizers have been confirmed, because the raw material was perfectly clinkered at low temperatures of 1350°C, this is about 200°C less than the clinkering temperatures of traditional white cement raw mixes.

Table 5 – Composition of mineralizers in raw mixes (Raina and Janakiraman, 1998)

Raw mix	$CaF_2/ CaSO_4$ (%)
RM-1	-
RM-2	0.5
RM-3	1.00
RM-4	1.00*
RM-5	0.5 +1.0*
RM-6	1.0 +1.0*

* as SO_3

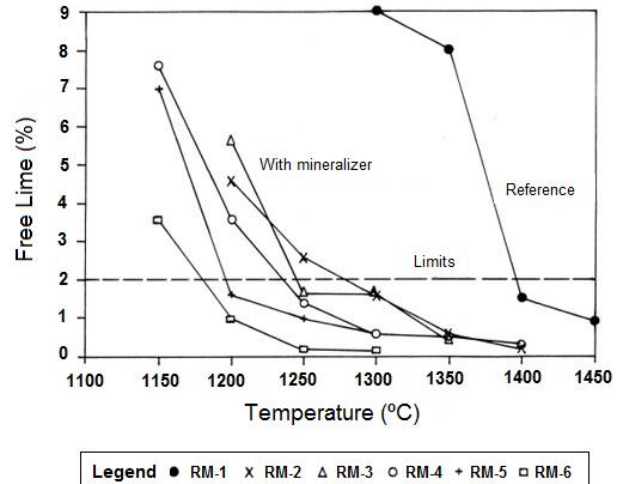


Figure 2 – Free lime at different temperatures for raw mix (Raina and Janakiraman, 1998)

Raina and Janakiraman (1998) also studied the use of CaF_2 and $CaSO_4$ in clinker production with main to promote the clinker formation at lower temperature.

The work used raw material from VSK (Vertical shaft kiln) cement plant for preparing raw mixes. Natural gypsum ($CaSO_4$) and fluorspar (CaF_2) were used.

The composition of mineralizers in raw mixes is present in Table 5. The results can be observed in Fig 2.

The changes of burning temperature of Portland clinker are presented in Fig (2). In this case it is expressed by the free lime content, caused by the presence of mineralizer mixed with raw mix. A free lime value of less than 2%, which corresponds to a well-burnt clinker, was obtained by burning RM-1 between 1400-1450°C. After using mineralizer (CaF_2 and $CaSO_4$) in the raw mixes (RM-2 to RM-6) the free lime content decreases substantially and the value of less than 2% can be achieved at temperature range 1300 – 1350°C. The main reasons are due to:

- The decomposition of $CaCO_3$ occurs above 1000°C; this process absorbs a large amount of heat. The presence of SO_3 and F- speeds up $CaCO_3$ decomposition at a lower temperature, thus reducing heat consumption or fuel inputs.
- The dissolution of C_2S and CaO ion diffusion accelerates C_3S formation. The crystal growth of C_3S is also favored by the liquid phase formation at lower temperature. At the same time SO_3 is able to stabilize – C_2S , preventing.

Other mineralizer known by cement industries is the phosphogypsum that is a by-product of the fertilizers

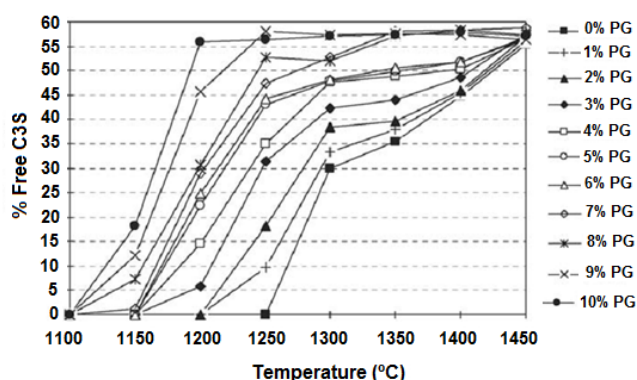


Figure 3 – Phosphogypsum effect on C₃S formation at different temperatures (Kacimi *et al.*, 2006a).

industry, obtained by etching natural calcium phosphates by sulfuric acid. Kacimi *et al.* (2006a) analyzed the valorization of phosphogypsum in the cement production, as well as its influence on the burning temperature of clinker and hydraulic properties of cement.

The work observed the variation of the formed alite (C₃S) content in the mixtures containing different phosphogypsum versus burning temperature (Fig.3).

From Fig. 3, it is presented that the increase of phosphogypsum (PG) content in the mixture cause quickly increases of the alite content at burning temperature. This is due to the catalytic effect of the waste, which accelerates the lime combination process and leads to the alite formation at low temperature.

The temperature of alite complete formation, which is clinker formation temperature, decreases versus the phosphogypsum content. Thus, for 10% phosphogypsum, the clinker contains 56% alite at low burning temperature (1200°C).

The structural and morphological analysis of the clinker, produced under these conditions, showed that this mineralizer preserves, at low burning temperature (1200°C) the silicate phases crystallinity usually acquired at high temperatures (higher than 1450°C). The results also appointed that the low amorphous phase amount in this clinker increases its porosity and consequently, facilitates its grinding and improves its specific area. This leads to the improvement of its hydraulic properties.

Taher (2007) also investigated also the effect of mineralizer phosphogypsum in cement production by mixing Portland cement clinker and blast furnace slag with thermally treated phosphogypsum at different temperatures. The characteristics of prepared mortars were investigated by determination of compressive strength, bulk density and total porosity. The results show that Portland slag cement produced using 6% thermally treated phosphogypsum at 800°C improves its hydraulic properties.

Kacimi *et al.* (2006b) analyzed the influence of alkaline fluorides (CaF₂, NaF and KF) on the clinker burning temperature and its properties are described. Three different raw materials were used and were burned with

1% of these fluorides, and physical and mechanical properties of some samples were determined.

The results showed that the addition of fluorides to the clinker induced a decrease in the burning temperature (1300°C), while improving the clinker phase formation and its hydraulic properties. CaF₂ seems to reduce the hydraulic activity; NaF improves the clinker crystallization at low temperature, by supporting massive formation of alite. Thus, the hydraulic activity is increased, and the physical and mechanical properties of the clinker are improved. The KF improves the product resistance; ensure by presence of alite that increases the hydraulic activity due to its disturbed crystalline structure.

Benarchid and Rogez (2005) studied the influence of chromium and phosphorous additions on the phase transformations during the formation of the calcium sulfoaluminate from CaCO₃ – Al₂O₃ – CaSO₄ mixtures. The results show that the Cr₂O₃ and P₂O₅ additions (0.31 and 0.58 wt% respectively) lower the onset temperature of decarbonation (1075°C). The optimal measure value of enthalpy was 1594 J/g.

VI. CONCLUSIONS

The valorization of the polluting waste enables to contribute to the improvement of ecological balance and the environmental protection by its recovery and incineration in the production of clinker Portland cement.

The decrease of clinker burning temperature caused with the application of wastes as mineralizers can reduce the pollutants emissions from cement industry due to reduction of fuel consumption.

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