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NO_x FORMATION IN CO-PROCESSING OF USELESS TIRES IN THE CEMENT INDUSTRY CLINKER KILNS

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Abstract: *Much industrial waste can be co-processed in the clinker kilns, resulting in their thermal destruction as well as taking advantage of their net calorific value in the process. The useless tires are employed in the co-processing in the clinker kilns as an alternative fuel, allowing a great deal of destruction. The burn of these used tires represents a discard alternative solution that constitutes an environmental problem as well as replacing the use of fossil fuels. This work analyzes the* NO_x *formation process inside the clinker kiln with the burn of the useless tires along with petcoke. In the burn process are analyzed: the feeding tires rate in the kiln, the scrap tires dimensions, the local kiln introduction, the temperature, the air/fuel ratio and others influential factors. Through a simultaneous reactions model, the factors that decrease the* NO_x *emission in the kiln that co-process the tires and the possible levels of substitution of the main fuels are analyzed. The analyses still take account the burn being done in a rotary kiln endowed with cyclone, a kiln with preheater and precalciner (with secondary fuel burn), and if there is or not a cooled clinker system grid with tertiary air supply.*

Keywords: *Emissions of* NO_x*, Tires, Cement industry, Co-processing of residues.*

1. Introduction

To date, the high degree of the industrial activity and consequently increase of society quality of live have been accompanied by a growing generation of residues, which represent one of the most serious environmental problems. The restrictions imposed for the discard of these residues become more and more severe, demanding technological and economically feasible solutions to destine thousands of tons generated annually.

The cement system production employs a significant amount of fossil fuels as the main thermal source of energy. The replacement of part of this conventional fuel to alternatives ones in the world cement industry has been getting the attention of the scientific and industrial community of this sector.

Besides the economical advantages due to the cost reduction, it can be pointed out that most of the residues resulting from the burn are incorporated into the clinker, minimizing the pollutant emissions.

So, the cement industry can contribute significantly for the solution of a problem that affects most of the industrialized countries, that is, the continuous growth of residues that has to be disposed appropriately, in which the tires stand out.

Brazil produced about 45 million tires in 2003 (CEMPRE, 2004), which one third is exported for 85 countries and the remaining are employed in national vehicles. Despite of the high tire reuse index in Brazil, which extends its life around 40%, most of them already consumed by the use ends up in landfills, in the edge of rivers and highways, and even in houses backyards, where water is accumulated attracting insects that transmit diseases.

To date in Brazil, according to IPT researchers, 10% of the tires are recycled, which 57% are destined to the cement kilns. In the USA, according to EPA (2003), there are at least 300 million useless tires in stocks. Only in 2001, 281 million of useless tires were generated and 73% were destined to the cement kilns (CEMPRE, 2004).

Brazil Environment National Council - CONAMA - has been imposing severe laws to the tires manufacturer regarding to the tires discard after their useful life through the resolution number 258, of August 26, 1999, which establishes the tires manufactures and importers to withdraw the used tires in order to reduce the environmental impact. On the other hand, the control of the cement industries pollutant emissions has also been deserving attention according to the resolution number 264 of August 26, 1999, which in its section VII article 28 deals with the maximum atmospheric emissions limits when the co-processing of residues in clinker kilns takes place. So, the burn of the tires in the cement kilns seems to be an available alternative for both, tires and cement industries.

This work analyzes the NO_x formation process inside the clinker kiln with the burn of the useless tires along with the petcoke. In the burn process are analyzed: the feeding tires rate in the kiln, the scrap tires dimensions, the local kiln introduction, the temperature, the air/fuel ratio and other influential factors. Through a simultaneous reactions model, the factors that decrease the NO_x emission in the kiln that co-process the tires and the possible levels of substitution of the main fuels are analyzed. The analyses still take account of the installation type, i.e., the burn being done in a rotary kiln endowed with cyclone, a kiln with preheater and precalciner (with secondary fuel burn), and if there is or not a cooled clinker system grid with tertiary air supply.

2. NO_x formation literature review

There are usually three main pollutants species considered in the combustion processes: unburn hydrocarbon (UHC), carbon monoxide (CO), and oxides of nitrogen (NO and NO_2 usually call NO_x) (Richards *et al.*, 2001). Both UHC and CO are incomplete combustion products. With enough residence time, and high temperatures, these two pollutants will be rusted to carbon gas and water.

Attempts in the use of fuels of low calorific value demand a greater residence time to complete the oxidation.

During the combustion process the nitrogen of the air or contained in the fuel is converted to pollutant containing nitrogen such as, NO, NO_2, N_2O, NH_3 and HCN. The species of pollutants formed depend mainly on the temperature and the fuel/oxygen in the combustion area ratio.

The high temperatures involved in the cement production process in the rotary kilns benefits the oxides of nitrogen (NO_x) formation.

NO_2 formation tends to happen where there are fast cooling regions, for example, in combustion hot mixture region with air inlet.

Emissions of NO_x in the combustion systems result of three main processes: Thermal NO_x, Fuel NO_x and Prompt NO_x. In cement kilns the thermal and fuel NO_x are the most relevant processes.

Thermal NO_x is formed by the oxidation of the atmospheric nitrogen. Fuel NO_x is formed by the oxidation of the nitrogen compounds in the fuel. Prompt NO_x is formed by the reaction of the atmospheric oxygen with the hidrocarbons radicals in the flame front area.

Thermal NO_x is the main source of NO_x in gaseous combustion systems and fuel NO_x is the main source of NO_x in coal burn systems.

NO_x emissions evaluation is quite complex and the understanding of the combustion models and the NO_x reaction processes are important tools that can be used in the study of the pollutant processes formation.

2.1. Thermal NO_x formation

Thermal NO_x is the main route of the mechanism of NO_x emissions formation in the cement production due to the high temperatures involved in the burn or in the clinker formation stages along with the kiln atmosphere oxidizer which contribute to a great amount of NO_x formation. The factors that contribute to thermal NO_x formation are: the gaseous temperature phase, concentrations and the contact between the nitrogen and oxygen.

The oxidation of NO in NO_2 takes place in the kiln low temperature regions. However the NO_2 usually appears in percentages smaller than 10% of the NO_x emitted by an exhausted system of a rotary kiln (U.S. EPA, 1994 and Silva, 1994).

Thermal NO_x is formed by a homogenous reaction of oxygen and nitrogen in the gaseous phase at high temperatures. Zeldovich, a Russian researcher, was the first who proposed a mechanism for the NO_x formation. Its formation consists of certain reaction stages as described by Eq. (1) to Eq. (3) (Miller and Bowman, 1989 and Van der Lans *et al.*, 1997).

$$N_2 + O \leftrightarrow NO + N \tag{1}$$

$$N + O_2 \leftrightarrow NO + O \tag{2}$$

$$N + OH \leftrightarrow NO + H \tag{3}$$

The thermal route is a primary mechanism for a NO_x formation when the temperature of the flame is approximately above 1800 K. Below this temperature, the thermal reactions are relatively slow and thermal NO_x formation is significantly reduced. The NO_x formation is also very smaller in the central zone region of the flame due to the low concentration of the oxygen atoms (Hill and Smoot, 2000).

The literature has demonstrated through an experimental study that thermal NO_x concentration grows linearly with the atomic oxygen concentration and exponentially with the temperature (U.S. EPA 1998).

2.2. Fuel NO_x formation

Fuel NO_x is formed during the combustion by the nitrogen oxidation contained in the fuel usually through the HCN formation and/or NH_3 that oxidized to NO being forwarded reduced to N_2 according to the reactions described by Eq. (4) and Eq. (5) (Hill and Smoot, 2000).

$$HCN / NH_3 + O_2 \rightarrow O_2 \rightarrow NO + ... \tag{4}$$

$$NO + HCN / NH_3 \rightarrow N_2 + ... \qquad (5)$$

Nitrogen contained in the fuel is liberated during the devolatilization process. A fraction of the nitrogen is rapidly converted to HCN, and the remaining portion of the fuel nitrogen reacts to form NH_3. These two species also react to form either NO or N_2 depending on the local conditions, that is, in fuel-rich areas (high concentration of nitrogen) the species containing nitrogen will be reduced to N_2, and in fuel-lean conditions (low concentration of nitrogen) the species will be oxidized to form NO. So, one way to control the NO_x emissions is to control the local environment in which the nitrogen is liberated from the fuel.

Fuel NO_x is formed more rapidly than the thermal NO_x due to N-H and N-C bonds most common in fuel-bound nitrogen are much weaker than the triple bond in molecular nitrogen which must be broken for thermal NO_x formation. (Hill and Smoot, 2000)

Among the factors that influence fuel NO_x formation are: the burner geometry, contact techniques between the fuel and the air, high rates of mixtures, nitrogen concentration in the fuel, volatilization rate, fuel burn and gaseous temperature.

The effect of air/fuel mixture tends to decrease when the mixture becomes richer in the fuel. CO also influences the process of NO_x reduction through the radicals (OH, H and O) formed by the CO oxidation (Hill and Smoot, 2000).

Coal, natural gas, oil, chemical solvents residues, tires, etc. have been using more and more by the cement industry. Nitrogen percentage contained in these fuels is very diverse. Depending on the source the coal can contain 1-3% of nitrogen in weight. The residues derived fuels such the organic residues can contain nitrogen in significant concentrations depending on the chemical species contained in the fuel to be burned.

Employing coal as a primary fuel and with 10% conversion of coal nitrogen to NO_x, 0.7 kg of NO_x (expressed as NO_2) may be formed per ton of clinker (U.S. EPA, 1994).

Due to the difficulty of identifying fuel NO_x from the thermal NO_x, the measures of the concentrations indicate only the total NO_x formed in the process. Fuel NO_x starts to form between 900-1000 K, and at 1600 K they can constitute about 70-80% of the total NO_x emission (U.S. EPA 1998).

2.3. NO_x prompt formation

These oxides of nitrogen receive this denomination due to their high-speed formation in the flame front region.

Fenimore (1971) was the first who studied the prompt NO_x formation mechanism in which the atmospheric nitrogen reacts with the hydrocarbon radicals being later oxidized forming NO_x.

The main reactions proposed for this mechanism are described by Eq. (6) to Eq. (8).

$$N_2 + CH_x \leftrightarrow HCN + N + ... \qquad (6)$$

$$N_2 + C_2 \leftrightarrow 2CN \qquad (7)$$

$$N + OH \leftrightarrow NO + H \qquad (8)$$

Eq. (6) is the main reaction in the NO_x formation mechanism. It is considered that approximately 90% of HCN participate in the prompt NO_x formation.

Prompt NO_x formation takes place in the flame front region where the hydrocarbon radicals increase the HCN formation according to the Eq. (9) and Eq. (10).

$$CH + N_2 \leftrightarrow HCN + N \qquad (9)$$

$$CH_2 + N_2 \leftrightarrow HCN + NH \qquad (10)$$

Prompt NO_x formation mechanism is more predominant in the fuel flames rich in hydrocarbon that in the flames of deficient fuels in hydrocarbon (Hill and Smoot, 2000).

Other characteristics of fast oxidation are: brief duration of the process, weak dependence among the NO_x emission and the temperature and strong dependence of the NO_x emission with the air/fuel ratio (Hill and Smoot, 2000).

Prompt NO_x reactions are neglected in many NO_x models due to the increased complexity of the nitrogen chemistry and also due to intimate coupling of these reactions with the fuel oxidation steps (Hill and Smoot, 2000).

3. NO_x formation in clinker kilns of cement industries

3.1. Preheating suspension kiln system with riser duct firing

According to Nielsen and Jespen (1991), in many preheating suspension kiln systems, 10-20% of the fuel is fired into the riser duct reducing NO_x emission. This reduction is probably explained by the fact that a large part of the fuel directly falls down into the kiln charge, creating a reducing atmosphere in the bottom part of the kiln back end in which NO_x from the burning zone is reduced.

Conversely, when firing finely ground fuel into the kiln riser duct, the specific NO_x content in the exhaust gas will often increase on passing through the riser duct. As NO_x emission from the kiln may also increase slightly due to an increased excess air rate, the total NO_x emission from the kiln system will often increase when starting up riser duct firing with finely ground fuel.

3.2. Precalcining kiln systems

The precalciner employs around 60% of the total fuel demand for clinker production in a secondary firing system, which reduces to 40% the requirements for the primary burner in the rotary kiln. The fuel combustion in the secondary burn region supplies the necessary heat for the endothermic calcination of the calcium carbonate ($CaCO_3$) given by Eq. (11).

$$CaCO_3(s) \rightarrow CaO(s) + CO_2(g) \tag{11}$$

The raw yet calcined is taken by the gas flow to the last cyclone where it is unloaded. In precalcining kiln systems with tertiary air duct, firing into the rotary kiln typically accounts for only 40 to 50 percent of the total heat consumption and the specific amount of combustion gases from the kiln burning zone is reduced proportionally. On the other hand, the NO_x concentration in the kiln gas may be considerably higher than in preheater kilns.

This is probably due to the shorter material and longer gas retention time in the precalciner kiln burning zone combined with a very high secondary air temperature

There are basically two kinds of kiln system with precalciner, namely:

In-Line Calciner Systems (ILC): In these systems, the fuel combustion in the calciner takes place in a mixture of kiln exhaust gas and hot air from the cooler (tertiary air). Some of the nitrogen in the fuel reacts with NO_x from the kiln exhaust gas, whereas another part reacts with oxygen (from the tertiary air) to form NO_x. In most cases, the calciner contributes a little to the NO_x emission

Separate Calciner Systems (SLC): In these systems, the combustion in the calciner takes place in pure hot air. The NO_x in the calciner exhaust gas is added to the NO_x in the gas from the rotary kiln, which leaves this type of kiln system without being reduced. When fired with solid fuels, SLC systems must therefore be expected to generate somewhat higher NO_x emissions than ILC systems.

The change in the calcination burn regions and calcination reaction, that is, the precalciners use, offers several and significant advantages in which we can mention: the rotary kiln capacity double; the limestone consumption is four times smaller; reduction in the precalciner combustion temperatures (< 1373 K) with a consequent NO_x emission decrease and stability and control improvement in the rotary kiln.

4. The reburning principle (fuel staging)

In this method, besides the primary combustion in the rotary kiln (clinker formation around 1473-1753 K), it happens a secondary combustion in the precalciner, to a lower temperature (873-1173 K) with a supply of combustion tertiary air through an external duct to the rotary kiln. This combustion has the purpose of completing the preheating and calcination of the raw. Fig. 1 shows a schematic of the reburning process in a precalciner cement kiln.

The method includes the preheating/precalciner use, in which usually use tire scraps or shred, blots oily, solvents residues, etc. as complementary fuel.

The reburning process in a precalciner can be divided into three zones; the primary zone (rotary kiln), the reburning zone and the burnout zone.

In the reburning zone takes place the secondary fuel combustion (a mixture containing 20% of tire and 40% petcoke) with the rotary kiln exhausted gas and hot tertiary air around 1237 K coming from the clinker cooler.

The main mechanism of this reaction type is described by the CH_i radical formation originating from the reburning fuel combustion. NO_x formed in the rotary kiln primary combustor kiln reacts rapidly with CH_i, converting it partially in HCN, NH_3 or N_2, as shown by Eq. (12) to Eq. (14).

$$Fuel.... \rightarrow CH_i + ... \tag{12}$$

$$CH_i + NO \rightarrow XN + ...\qquad(13)$$

$$XN + NO \rightarrow N_2 + ...\qquad(14)$$

where $XN = HCN, NH_3$ ou N_2.

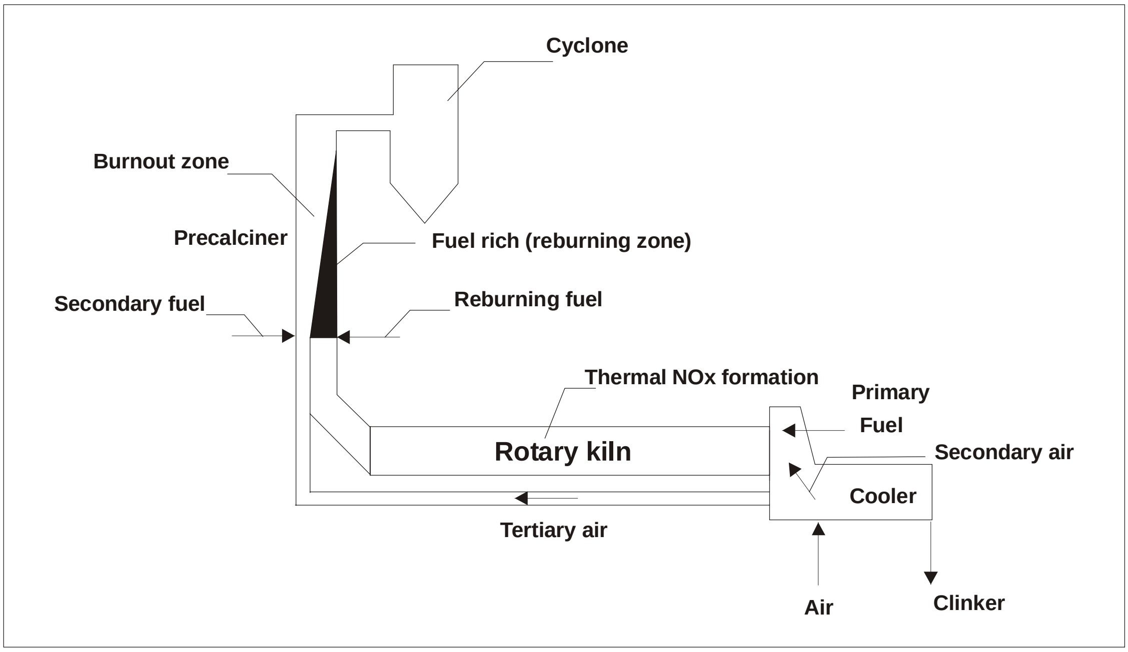

Figure 1. The reburning process in a precalciner cement kiln

The excess of oxygen supplied in the exit region of the burning gases reacts with the nitrogenous species (XN) to form nitric oxide or reduced to molecular nitrogen, Eq. (15) and Eq. (16) respectively. This is one of the reasons why reburning reactions can be efficient even at high oxygen levels in the kiln inlet.

$$XN + ... \rightarrow NO + ...\qquad(15)$$

$$XN + ... \rightarrow NO_2 + ...\qquad(16)$$

In the next phase, most of the oxygen is used and HCN reacts with OH creating NH_3. These NH_3 reduces NO_x via catalysis usually presents as iron oxides in the raw meal (Syverud *et al.,* 1994).

The reburning fuel benefits the reduction of the thermal NO_x formation in the secondary burning region due to the lower temperature. However, if the fuel used in the reburning fuel contains nitrogen in its composition, it can be formed fuel NO_x and other species of nitrogen in the zone under precalciner reducing atmosphere, following the same NO_x reduction route formed in the rotary kiln primary burner.

When solid fuels are used in the reburning fuel, a reaction of heterogeneous reduction can happen, Eq. (17) (Tokheim *et al.,* 1998).

$$NO + (-C-) \rightarrow (-CO) + \frac{1}{2}N_2\qquad(17)$$

The groups in parentheses are mainly in the form of soot and in the presence of NH_3 they can also catalyze the reduction reaction of NO_x.

The heterogeneous catalytic reaction NO-CO can also happens according to Eq. (18) giving an additional contribution to the NO_x formation decrease.

$$NO + CO \rightarrow CO_2 + \frac{1}{2}N_2\qquad(18)$$

The reburning fuel is influenced by several process parameters, such as: temperature, air/fuel ratio, residence time in the reburning zone, through the fuel mixture type and conditions used and according to calciner geometry which consequently affects the NO_x formation processes.

According to Tokheim *et al.* (1998), all these processes parameters can be manipulated in the calciner. Varying the location and rate of the reburning fuel in which the fuel is burned, it can be controlled the stoichiometry, the residence time and the temperature in the burn secondary zone.

With the reduction of the air primary ratio one can usually gets a decrease in NO_x formation because it reduces the amount of available oxygen to react with the nitrogen and others nitrogenous compounds in the temperature peak vicinities. On the other hand, in general, an increase in the secondary air causes an increase in NO_x formation.

The mixture process can also be influenced by the fuel particles size and the way that it is added in the calciner.

The particles size influences the heating rate and temperature peacks, in other words, smaller particles hold a fast heating rate reaching high temperature peaks resulting in a larger concentration of volatile containing nitrogen in the gases, so contributing this way to fuel NO_x formation (Courtemanche, 1998 and Hill and Smoot, 2000).

Fuel derivate tire (TDF) fed at a rate that does not compromise metals input/output balances, has no effect on clinker quality. Stack emissions of CO, however, may be affected due to the manner in which the tires are fed into the kiln. A uniform feed rate of scrap or shred will allow the operator to increase the kiln exit oxygen, or rather allows the operator to maintain the desired kiln exit oxygen concentration and/or the kiln exit CO concentration. However, the insertion of whole tires at one to two minute intervals will often produce a CO spike and/or an oxygen dip in the kiln exit gases. This can be compensated by increasing the interval between tire insertions and/or by increasing the normal kiln exit oxygen by 0.5 to 2% points (Constans and Gossman, 1997)

5. Simultaneous Reactions

Aiming to evaluate the behavior of the tire and petcoke burn in the process of NO_x formation inside the kiln for clinker production, flame temperature values were evaluated for 34.72 kg/s clinker production considering the elementary chemical compounds of these fuels, presented in Tab.1.

Table 1. Characteristics of the fuels used in the cement industry.

Fuel Elementar Composition	Petcoke(1) (%)	Tire(2) (%)
C	88,4	72,15
H	3,7	6,74
O	0	9,67
S	1,56	1,23
N	1,56	0,36
PCI (kJ/kg)	34350	32580

Source: (1) (Commadré and Salvador, 2005)
(2) Amari *et al.* (1999)

The values for the flame temperature were obtained applying Eq. (19) (Van Wylen and Sontag, 2003):

$$H_r = H_p \tag{19}$$

where

$$H_r = \sum_r n_e \times \left(h_f^0 + \bar{\Delta} h \right)_e \tag{20}$$

$$H_p = \sum_p n_s \times \left(h_f^0 + \bar{\Delta} h \right)_s \tag{21}$$

H_r and H_p refer to the reagents and products enthalpies respectively, n_e and n_s the kmol number of each reagents and products compounds.

The flame temperature in this work considering 2% of free O_2 in the primary burner of the rotary kiln was of 2229K.

In the secondary burner of the precalciner the flame temperature was of and 1161K with 1,7% of free O_2.

With the purpose of evaluating the contribution of the NO-CO reaction given by Eq. (18) in the NO_x mechanism, the Gibbs free energy and the thermodynamic equilibrium constant, Eq. (22) and (23), were evaluated based on the flame temperature. Later, it was evaluated the respective molars fractions in the compounds chemical equilibrium of Eq. (18), presented in Tab. 2. Finally, the reaction progress degree represented by the *x* variable in Eq. (24) was evaluated indicating the reagents conversion in products in a chemical reaction.

$$\Delta G(T) = \Delta H - T\Delta S \tag{22}$$

$$K = exp\left(\frac{-\Delta G}{RT}\right) \tag{23}$$

where
ΔG : Gibbs free energy;
ΔH and ΔS : enthalpy and entropy variation respectively;
K: thermodynamic equilibrium constant:
R: gas universal constant
T: system absolute temperature.

Table 2. Mass balance for equation (18).

Compounds	Start	Equilibrium	Molar Fraction
NO	1	1 - x	(1 – x)/(2 – 0.5x)
CO	1	1 – x	(1 – x)/(2 – 0.5x)
CO_2	0	x	x/(2 – 0.5x)
$0.5N_2$	0	0.5x	0.5x/(2 – 0.5x)
Total		2 – 0.5x	

$$K = \frac{\left(\frac{x}{2-0.5x}\right)\cdot\left(\frac{0.5x}{2-0.5x}\right)^{0.5}}{\left(\frac{1-x}{2-0.5x}\right)^{2}} \tag{24}$$

It was observed that the chemical reaction, represented by Eq. (18) in the equilibrium, has shown a tendency to move more in the direction of products formation ($k = 3.857x10^{11}$ and $x = 0.999999$), that is, NO reduced to molecular nitrogen and CO oxidized to CO_2, so reducing the NO_x formation according to the arguments discussed in this work.

6. Conclusion

The group of considerations employed in this work has shown that the NO_x emited by a clinker kiln is intimately related to the combustion process that happens in it.

The burn of useless tires besides the environmental benefit and partial replacement of fossil fuels employed in the cement industry, also seems not to bring about adverse effects in the clinker quality and NO_x emissions when burned until approximately 20%.

It is believed that the fuel reburning is the main mechanism for NO_x reduction when tires are co-processed in kilns for clinker production.

The present study confirmed that the results obtained in measurements accomplished in cement industry clinker kilns that begin to employ useless tires co-processing regularly as alternative fuels presented NO_x reduction according to Baumhardt (2004). The measurements revealed reductions of emissions around 900 mg/Nm^3 to 11% of free O_2 for values above 300 mg/Nm^3. Therefore, it was verified that the burn of tires promotes the CO generation and the heterogeneous catalytic reaction NO-CO contributes to the decrease of NO_x formation in the process, object of this study.

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