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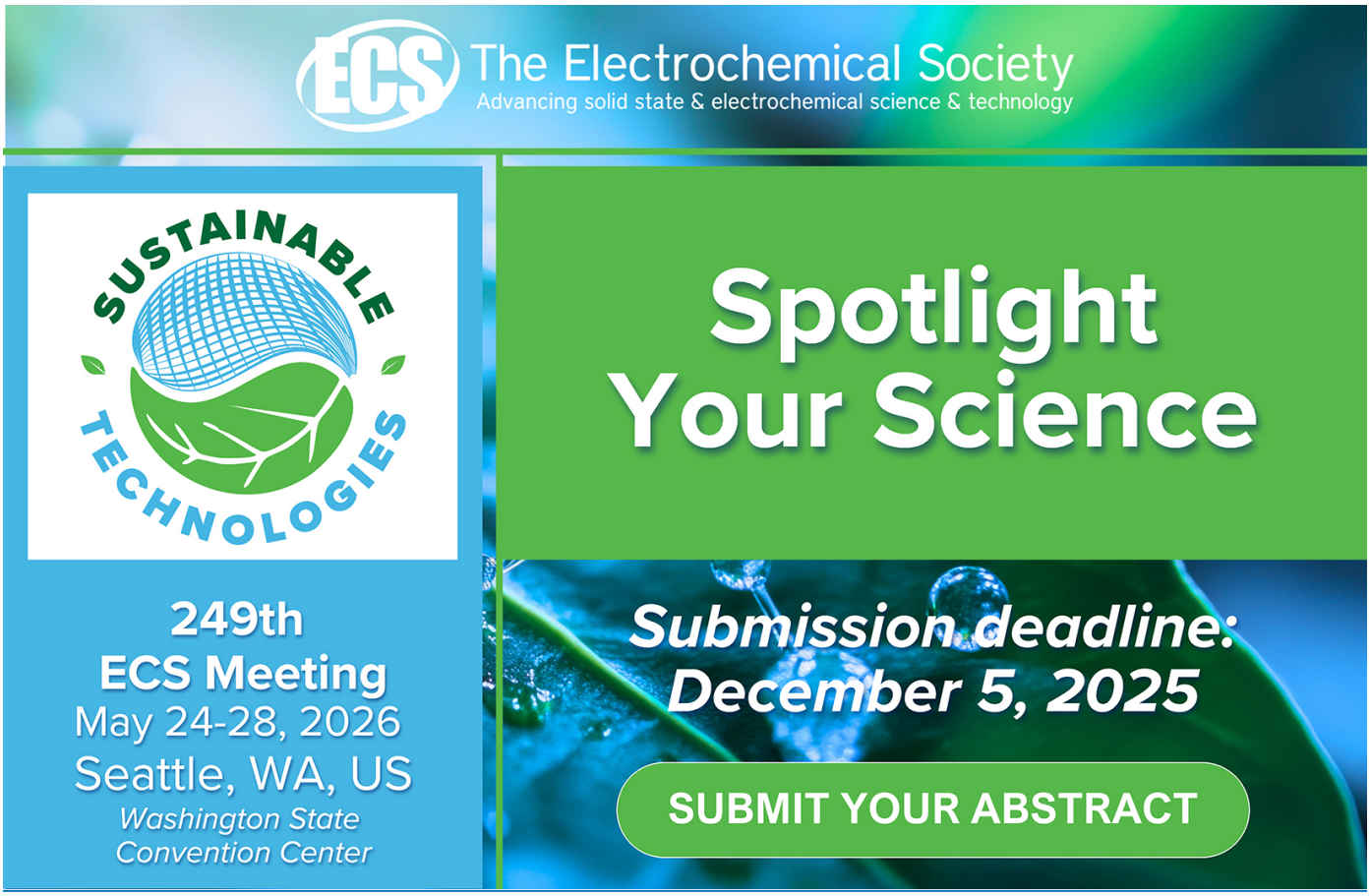
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Analysis of pollutant emissions using calcium carbonate in the manufacture of ceramic materials

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Abstract. In the research work, an isokinetic sampling of the evaluation of pollutant emissions to the atmosphere generated by a Hoffman continuous kiln was carried out in samples with two dosages, one composed of 100% clay and the other with 70% clay and 30% calcium carbonate. The methods for the evaluation of pollutant emissions in stationary sources were used and 3 runs were performed, and in each run 4 measurements were made every 15 minutes. The results in the emissions of the firing using calcium carbonate with respect to the samples composed of 100% clay reduced the discharges to the atmosphere by 7.30% in particulate matter, 20.18% in nitrogen oxides, 21.65% in sulfur dioxide, and 15.35% and 8.89% in hydrochloric and hydrofluoric acid respectively. The concentration of particulate material emitted to the environment in the two firing processes does not meet the permissible emission standard of less than 250 mg/mm^3, while the other pollutants comply with Colombian environmental regulations. The use of fluxes for the manufacture of bricks reduced the concentrations of gases emitted into the environment decreasing possible health effects such as acute respiratory disease, eye irritation, and aggravation of heart disease.

1. Introduction

The ceramic industry is one of the largest polluters of the environment and human health [1], causing environmental effects such as smog [2], acid rain [3], greenhouse effect [4], and weakening of the ozone layer [5]. The changes in the environment are presented by the extraction of raw materials [6], the consumption of fossil fuels and the poor control of combustion processes; therefore, it is necessary to use more efficient kilns that lead to a complete combustion [7] of the firing process and the use of new raw materials [8].

In Colombia, measures were adopted to counteract the effects of polluting gases emitted by the different industries and the ceramic industry of the country. The methodologies of monitoring [9] and evaluation of stationary sources of emission of atmospheric pollutants established by the environmental protection agency of the United States of America and the Kyoto Protocol were implemented [10]. The use of fluxes such as calcium carbonate in the composition of the ceramic material changes the mineralogical properties of the clay[11]. This can lead to a reduction in parameters such as firing time, sintering temperature, fuel consumption, temperature of gases released into the environment[12], and emissions of gases into the environment[13].

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The objective of the research is to demonstrate the influence of calcium carbonate to mitigate the environmental impact generated by the kilns used for the production of ceramic materials. For this purpose, an isokinetic sampling of the evaluation of the discharge of pollutants into the atmosphere of two brick dosages was carried out according to the regulations established by Ministerio de Ambiente y Desarrollo Sostenible, Colombia [14].

2. Methodology

This study is the continuation of the work use of calcium carbonate as a flux in the production of ceramic bricks in Ocaña, Norte de Santander, Colombia. The firing process was carried out in a Hoffmann continuous kiln in which two solid bricks batches were fired. In the first firing, the samples were made of 100% clay, and 1.500 kg of coal, with a production of 4.300 bricks. The firing process lasted two days and nine hours. In the second firing, the samples were made of 70% clay, 30% calcium carbonate, and 1.370 kg of coal, with a production of 4.300 bricks. The firing process lasted two days and three hours. The clay used to make the two sample compositions is from Algodonal, Colombia, intrusive and extrusive igneous formation.

The kiln is made up of 22 firing chambers, in each chamber 5 piles of bricks are loaded (see Figure 1), and each pile is made up of an average of 600 bricks [15]. The kiln has an average monthly production of 562.200 units and a coal consumption of 205.260 kg/month [16]. The coal is pulverized and injected through the upper part of the kiln using a carbojet. The first study determined the reduction in the time of the firing process, the fuel consumption per brick, the firing temperatures of the products, the gases released into the environment, and the mechanical properties of the products. This second study presents the emissions into the environment of the different polluting gases from the two firing processes of compositions mentioned above. This is carried out in accordance with [14].

2.1. Procedure for the evaluation of pollutant emissions

A previous visit to the brickworks was made to know the source of emission, in which the characteristics of the chimney such as height, as well as the locations and monitoring points in the chimney, were verified. The concentration of nitrogen oxides, particulate matter, sulfur dioxide, hydrofluoric and hydrochloric acids were measured using a Bacharach combustion gas analyzer for the firing process of 100% clay, and 70% clay and 30% calcium carbonate dosages. To know the characteristics of the emissions in the chimney, a preliminary sampling was made according to the methodology and procedures of the standards for air emissions established in methods 1, 2, 3, and 4 of the Environmental Protection Agency of the United States of America for the monitoring of pollutants discharged by stationary sources established in the protocol for the control and monitoring of air pollution generated by stationary sources. In the evaluation of pollutant emissions, the gas velocity, the concentrations of oxygen (O_2) and carbon dioxide (CO_2), the emission of particulate matter, the concentration of sulfur dioxide (SO_2), nitrogen oxides (NO_x), and the moisture content of the gas were determined. The measurement of gases in the chimney of the Hoffman kiln was performed in three pipes or nipples every 15 minutes for a total of three runs.

In the isokinetic sampling, a representative sample of the generating source was taken without altering the kinetic conditions of the same. For the collection of the sample, the condition of not generating the separation of the pollutants at the same speed with respect to the carrier gas was met.

2.1.1. Adjustment of pollutant measurement values to reference conditions. The measurements of the pollutants are adjusted according to the equations stipulated in the methods adopted by [14], thus obtaining the different variables for each run. In accordance by [14], an adjustment is made to reference conditions at 25 ºC and pressure of 760 mm. Hg in the results of the measurements of the pollutants, by means of the Equation (1).

$$C_{CR} = C_{CL} * \left(\frac{T_{CL} * P_{CR}}{T_{CR} * P_{CL}} \right), \quad (1)$$

where C_{CR} and C_{CL} are the pollutant concentrations at reference and local conditions respectively in (mg/m^3), T_{CL} and T_{CR} are the temperature of the gases at the duct outlet and at reference conditions in (K) and P_{CL} and P_{CR} are the pressure of the gases at the duct outlet and at reference conditions in (mm Hg). In accordance with Article 86 of Resolution 909 of June 5, 2008 issued by [14], to evaluate the concentration at local conditions, the following expression was used see in the Equation (2).

$$C_{CL} = C_{CL\ p.p.m.} * \frac{P.M.}{24.45}, \quad (2)$$

where $C_{CLp.p.m.}$ is the local concentration in parts per million and P.M. is the molecular weight of the pollutant in grams.

2.1.2. Adjustment of the reference values to oxygen conditions. According to article 88 of Resolution 909 of June 5, 2008 issued by [14], an adjustment to reference oxygen conditions of 18% is made using the following Equation (3).

$$C_{CR(O2\ ref)} = C_{CR\ (X\ \%)} * \left(\frac{21.00 - \%\ O_{2\ ref}}{21.00 - X\ \%}\right), \quad (3)$$

where $C_{CR\ (O2\ ref)}$ and $C_{CR\ (X\ \%)}$ are the pollutant concentration at reference conditions and 18% oxygen in (mg/m^3) and X % is the oxygen measured at the exit of the gases in (%).

3. Results

Tables 1 and Table 2 show the results obtained from the evaluation of the emissions generated from the Hoffman kiln for the two dosages in the three runs and their respective average.

Table 1. Emission of pollutants for samples with 100% clay dosage.

Concentration	Run 1	Run 2	Run 3	Average
Particulate matter (mg/m^3)	2454.8	2485.3	2550.6	2496.9
Nitrogen oxides (mg/m^3)	65.3	69.2	66.8	67.1
Sulfur dioxide (mg/m^3)	58.7	55.9	57.3	57.4
Hydrochloric acid (mg/m^3)	6.9	7.8	6.1	6.9
Hydrofluoric acid (mg/m^3)	5.4	6.3	4.8	5.5

Table 2. Emission of pollutants for samples with 70% calcium carbonate dosage.

Concentration	Run 1	Run 2	Run 3	Average
Particulate matter (mg/m^3)	2640.8	2589.5	2705.5	2645.8
Nitrogen oxides (mg/m^3)	114.9	119.3	108.4	114.2
Sulfur dioxide (mg/m^3)	53.8	52.1	55.1	53.7
Hydrochloric acid (mg/m^3)	6.5	4.4	9.5	6.9
Hydrofluoric acid (mg/m^3)	5.5	5.3	6.5	5.8

Table 3 and Table 4 show the emissions generated by the Hoffman kiln for the two dosages, under reference conditions of pressure and temperature. The results are compared with the admissible emission standards for air pollutants established in Articles 30 and 32 of Resolution 909 of 2008 of the Ministerio de Ambiente, Vivienda y Desarrollo Territorial of Colombia [14]. The results for particulate matter for dosages containing 100% clay and calcium carbonate are 735 mg/m^3 and 685 mg/m^3 respectively at temperature and pressure reference conditions, and at an oxygen concentration of 18%. There is a 6.80% reduction in particulate matter emissions when using calcium carbonate over a 100% clay dosage. However, the results obtained are above the established limit 250 mg/m^3, in percentages of 294% and 274% with respect to the standard for dosages containing 100% clay and calcium carbonate respectively. Significant concentrations have been recorded that can affect air quality since there are no control

mechanisms to reduce the emission of this pollutant into the atmosphere; as a result, controls on companies in this area should be more rigorous.

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The results of nitrogen oxides for the dosages containing 100% clay and calcium carbonate are 137 mg/m^3 and 114 mg/m^3 respectively, obtaining a 16.79% reduction of emissions when using calcium carbonate with respect to a 100% clay dosage. However, the results obtained are below the established limit 550 mg/m^3, with an index of 401% and 482% with respect to the standard. The results of nitrogen oxides for the dosages containing 100% clay and calcium carbonate are 137 mg/m^3 and 114 mg/m^3 respectively, obtaining a 16.79% reduction of emissions when using calcium carbonate with respect to a 100% clay dosage. However, the results obtained are below the established limit 550 mg/m^3, presenting a reduction of with an index of 401% and 482% with respect to the standard. The main reason for the reduction of nitrogen oxides in the kiln is that the temperature of the gases released into the environment does not exceed 300 °C.

The average concentrations of sulfur dioxides are 163 mg/m^3 and 134 mg/m^3 for the dosages containing 100% clay and calcium carbonate representing a decrease of 17.79% when using the flux. These concentrations are below the permissible limit of 550 mg/m^3 according to the resolution for stationary sources.

Table 3. Gas emissions under reference conditions and permissible standard comparison for samples with 100% clay dosage.

Concentration	C_{CL}	C_{CR}	C_{CR} O_2 Ref	Standard
Particulate matter (mg/m^3)	2496.9	1225	735	250
Nitrogen oxides (mg/m^3)	67.1	228.3	137	550
Sulfur dioxide (mg/m^3)	57.4	271.7	163	550
Hydrochloric acid (mg/m^3)	6.9	18.8	11.3	40
Hydrofluoric acid (mg/m^3)	5.5	8.2	4.9	8

Table 4. Permissible gas emissions under reference conditions and standard comparison for samples with 70% calcium carbonate dosage.

Concentration	C_{CL}	C_{CR}	C_{CR} O_2 Ref	Standard
Particulate matter (mg/m^3)	2645.8	1141	685	250
Nitrogen oxides (mg/m^3)	114.2	342	114	550
Sulfur dioxide (mg/m^3)	53.7	223.3	134	550
Hydrochloric acid (mg/m^3)	6.9	16.3	9.8	40
Hydrofluoric acid (mg/m^3)	5.8	7.5	4.5	8

The results for hydrochloric and hydrofluoric acids for the 100% clay and calcium carbonate dosages are 11.30 mg/m^3 and 9.80 mg/m^3 and 4.90 mg/m^3 and 4.50 mg/m^3 respectively. There is a 13.27% and 8.16% reduction in hydrochloric and hydrofluoric acid emissions when using calcium carbonate over a 100% clay dosage. In comparison with studies carried out in the department of Norte de Santander, Colombia, in the municipalities of “Los Patios”, “Villa del Rosario”, and “Zulia”, the concentrations of particulate material averaged 200 mg/m^3, 150 mg/m^3, and 250 mg/m^3 respectively [6]. These concentrations are all below the ones found in the Hoffman kiln of 735 mg/m^3 and 685 mg/m^3 for dosages containing 100% clay and calcium carbonate. The concentrations of nitrogen oxides were 90 mg/m^3, 33 mg/m^3 and 32 mg/m^3 respectively [6], which were below the concentrations found in the

Hoffman kiln of 137 mg/m^3 and 114 mg/m^3 for dosages containing 100% clay and flux. The concentrations of sulfur oxide were 379 mg/m^3, 221 mg/m^3, and 205 mg/m^3 respectively [6], which are above the concentrations found in the Hoffman kiln of 163 mg/m^3 and 134 mg/m^3 for dosages containing 100% clay and calcium carbonate. The isokinetic monitoring carried out in the kilns in the department of Norte de Santander, Colombia, are below the limit values as established in Resolution 909 of 2008 of Ministerio de Ambiente, Vivienda y Desarrollo Territorial of Colombia [14].

The average emissions of nitrogen oxides measured in the companies monitored in the department of Norte de Santander are low due to the outlet temperature of the gases, which at the time of sampling ranged from 55 °C to 150 °C [6], and in the dosages containing 100% clay and calcium carbonate was 225 °C to 165 °C.

4. Conclusions

The emissions generated by the Hoffman kiln during the firing of samples using calcium carbonate decreased the concentrations of particulate matter in the atmosphere from 735 mg/m^3 in the firing of samples using 100% clay to 685 mg/m^3. Both concentrations may generate adverse effects on human health since these results are above the limit of 250 mg/m^3 as established in of Ministerio de Ambiente, Vivienda y Desarrollo Territorial of Colombia The concentrations obtained of particulate matter determine a moderate level of pollution, generating possible health effects such as irritation to the respiratory tract and eyes, and possible deterioration of heart or lung disease in people with cardiopulmonary diseases. Possibly due to ash and soot carryover in the gases emitted due to a high excess air.

The concentrations determined of the pollutants nitrogen oxides, sulfur dioxide, hydrochloric acid and hydrofluoric acid during the firing of the two doses in the Hoffman kiln, are clearly below the standard of permissible emissions of pollutants into the air established in Articles 30 and 32 of Ministerio de Ambiente, Vivienda y Desarrollo Territorial of Colombia This reaffirms the reduction of air emissions generated by using fluxes for the manufacture of bricks, which is an environmentally sustainable, economically viable and socially acceptable alternative. The Hoffman kiln was found to be more efficient using calcium carbonate in the production of bricks, as it led to a 23.88% decrease in the time of the firing process, from 67 hours to 51 hours. On the other hand, a reduction in fuel consumption of 8.67% was achieved with total energy consumption from 11,441 kJ/kg brick to 10,450 kJ/kg brick, as well as a decrease in the temperature of the gases in the environment, registering a maximum temperature of 225°C to 165°C. Therefore, it can be inferred that the implementation of fluxes in the production of ceramic materials is a suitable option for the entrepreneurs of the ceramic sector.

According to Article 30 of Resolution 909 and Resolution 802 of Ministerio de Ambiente, Vivienda y Desarrollo. Territorial, Colombia, the emission temperature established in the exhaust gases in continuous kilns must be below 180 ºC. As a result, the gases released into the environment in the 100% clay dosage do not comply with the standard and the gases released into the environment using calcium carbonate comply with Colombian regulations. The height of the chimney does not meet the minimum required by resolution 1632 of 2012 of Ministerio de Ambiente, Vivienda y Desarrollo Territorial, Colombia, which must be 30 meters and the chimney of the kiln is 18.20 meters. As a result, the height of the chimney of the Hoffman kiln does not meet the standard.

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