Use of Non-Hazardous Solid Waste as Alternative Fuels in Cement Manufacturing Process: Impact on the Quality of Cement and Gas Emissions

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Abstract—The incineration of non-hazardous solid waste and its use as alternative fuel in cement manufacturing process was studied and simulated under the effect of air flow acceleration in a laboratory scale reactor. Firstly, analysis of the different waste materials (textile, wood and paper) was performed separately, showing that textile samples presented the highest levels of heavy metals (H.M). In the course of a test run using solid recovered fuel (SRF), the mass balance of heavy metals revealed that lead and chromium probably volatilized during firing while arsenic, cadmium and zinc were trapped in clinker. As to gaseous emissions, heavy metals concentration in the stack remained relatively low and below the standard limits. Secondly, the temperature and concentration of gases flue was monitored. It was shown that the combustion regime is characterized by low reaction temperatures and an oxygen-deficient environment. Air injection rate affected significantly the formation and degradation mechanisms of the emitted gases concentrations, particularly CO, CO₂, NO, NO_x, SO₂. Textile waste exhibited the lowest concentration of emitted gases compared to the other types of waste.

Index terms—Cement Kiln, Incineration, Non-Hazardous Solid Waste, Stack Emissions, Mass Balance of Heavy Metals.

I. INTRODUCTION

Waste incineration is a technology widely adopted due to its several advantages such as energy recovery and volume and mass reduction, since it has been technically proven as an ultimate solution for waste destined for landfill. However, it is known that waste incineration can cause air pollution, due to the emission of CO, CO₂, NO_x, SO₂ and NO which are polluting gases with known harmful effects on human health and the environment [1].

One of the best technologies adopted in solid waste recycling is their use as alternative fuel or as raw material substitute in cement kilns [2]. This is considered as a simultaneous way to eliminate residues, to reduce greenhouse gases and to produce energy. However, some wastes must be treated and used carefully in the cement kiln, especially those containing heavy metals and inorganic materials [3]. In fact, these components can be incorporated into the cement raw material (clinker) [1], [4], and then alter the quality of the cement and the emitted gases.

Several works were devoted to study waste incineration [5]–[7] under different combustion regimes and to examine the emission of pollutant gases. These studies allowed one to better understand the effect of different parameters on the

combustion mechanisms and the products generated throughout the process. In fact, according to earlier investigation, it was stated that the substitution of one ton of petroleum coke by one ton of solid waste fuels reduced CO₂ emissions by approximately one ton [8].

Furthermore, the concentration of the formed compounds depended strongly on the combustion regime. For instance, when the stoichiometric combustion conditions were fulfilled (i.e. the oxidizer amount was exactly enough to completely oxidize the fuel), the combustion was complete and only CO₂ and H₂O were the main combustion products. Nevertheless, in this case, the stoichiometric requirements were not satisfied, which resulted in the presence of unoxidized (unburnt) species. A lack of oxygen and low temperatures in the oxidizing medium were considered the primary reasons of such behavior.

Generally, an excess of injected air provided the necessary amount of oxygen, diluted the combustion fumes and drops the temperature of the oxidizing medium [7], [9]. The reactions are characterized by the air-fuel equivalence ratio λ as depicted in Fig. 1:

Fig. 1. Variation of air-fuel equivalence ratio λ .

The present paper considered the study of non-hazardous solid waste incineration under the effect of air acceleration at low temperature environment. These two parameters had an important effect on the nature and concentration of the emitted gases. The experiments were carried out by using a waste incinerator in which various combustion tests were conducted on three type of non-hazardous waste (textile, wood and paper). On one hand, a mass balance of heavy metals was established on representative samples of the waste, raw materials, pet coke, and clinker. On the other hand, the evolution of the combustion gases temperatures was estimated and the concentration of the principal polluting gases, namely O₂, CO, CO₂, NO, NO_x and SO₂ was measured.

II. EXPERIMENTAL PROCEDURE

A. Sampling method

In the case of solid waste, obtaining representative samples is quite a difficult and complicated task, due to the complexity and diversification of the waste composition

encountered in the field. Sampling process was conducted at a platform where non-hazardous and hazardous industrial wastes are collected and recycled. The deposits were chosen in a way to respect certain imperatives, such as the abundance of the collected waste, and to respond to cement manufacturer's specifications.

In the current study, we collected the following types of waste:

- Paper and cardboard waste: paper packaging, corrugated cardboard and flat packaging.
- Wood waste: sawdust, shavings and woodchip pieces that cannot be recycled as material.
- Textile waste: natural and synthetic fibers, and rags.

It is noteworthy to mention that a part of the collected waste originated from bulky waste deposited in the "all-in" skips bins, which consists generally of refuse sorting, scrap wood, plastic, cardboard, etc. Therefore, it was crucial to have a precise knowledge of the composition of these flows, which was why a selective sorting was performed in advance [10].

1) Samples preparation steps

The preparation of waste samples is conducted according to the following steps:

- Storage of incoming waste (under open shelter in order to promote drying and ensure proper aeration).
- Separation and manual sorting of heavy inert and unwanted materials (Metals, PVC).
- Waste sorting by typology.
- Shredding and grinding.
- Mixing and formulation of the fuel according to the desired composition [11].

2) Samples collection used for laboratory analysis

In order to prepare the laboratory sample, waste sampling standards must be followed. In this work, French sampling standards were taken as the reference. Generally, a procedure and 6 sampling guides used for a waste deposit are available [12]. The current sampling plan is based on the guidelines of these standards. The main steps of the sampling are summarized in Fig.2.

Fig.2. Principal steps of waste sampling according to French standards [12], [13].

B. Solid waste incineration

The equipment used to carry out the present work is a SmartAsh 100 incinerator. It consists of a cyclonic incinerator based on the principle of accelerating combustion under the effect of air injection. As it could be

seen in Fig.3, the air entered the combustion chamber through a hole in the upper part. The hole was positioned tangentially and slightly inclined downwards leading to the emergence of an air flow in the form of a cyclone. The acceleration of the air increased the temperature of the incinerated waste.

The experiments were performed on solid waste composed of paper and cardboard, rags and wood. In each test, the incinerator was loaded with the same amount of waste (10 kg). Incineration was triggered by the ignition of the test sample at several locations in the incinerator. Once the incineration began, the lid was placed underneath. The incoming air flow rate was measured using a flowmeter. Note that at the beginning of the experiment, the air flow was minimal, then it was adjusted to 50 Nm³/h.

Fig. 3. Schematic diagram of the SmartAsh 100 incinerator system.

C. Analysis of heavy metals and trace elements

Heavy metals (H.M) contained in the samples were determined by using atomic absorption spectrophotometer type 2000 purchased from Shimadzu [13].

1) Mass balance of input and output elements of cement production process

Seyler and al. [14] and Qizhong and al. [16] developed the following equations to calculate the mass balance of a cement kiln:

$$\Sigma m_{\text{input}} = \Sigma m_{\text{output}} \quad (1)$$

With:

$$\Sigma m_{\text{input}} = 0.35 m_{r.m.} + 0.0194 (m_{a.f.} + m_{f.f.}) + 0.32 m_{\text{air}} \quad (2)$$

$$\Sigma m_{\text{output}} = 0.231 m_{\text{clinker}} + 0.77 m_{\text{gas}} \quad (3)$$

Where:

m_{input} : Flow rate of the species total mass at the kiln entrance in kg/h.

$m_{r.m.}$: Flow rate of the raw material (flour) mass at the kiln entrance in kg/h.

$m_{a.f.}$: Flow rate of the alternative fuel mass at the kiln entrance in kg/h.

$m_{f.f.}$: Flow rate of the fossil fuel mass at the kiln entrance in kg/h.

m : Flow rate of the entering air mass in kg/h.

m_{output} : Flow rate of the species total mass at the kiln exit in kg/h.

m_{clinker} : Flow rate of the clinker mass at the kiln exit, in kg/h.

m_{gas} : Flow rate of the gas mass into the stack, in kg/h.

The input and output masses are weighted by process-dependent coefficients.

2) Mass balance of heavy metals in cement plant

In order to calculate the mass balance of heavy metals, a principle similar to the one stated above is applied. During the firing process, the heavy metals contained in fossil and alternative fuels combine with those comprised in raw material. The combustion reactions occurring at high temperatures (1400 °C in cement furnace) lead to the formation of volatile dusts and gases containing a percentage of heavy metals, which is added to the percentage of heavy metals remaining in the fired material [14], [15].

The overall mass balance of heavy metals should be calculated for each element individually. The material conservation equation for heavy metals is given as follows:

$$0.35 HM_{r,m} + 0.0194 (HM_{a,f} + HM_{f,f}) = 0.231 HM_{clinker} + 0.77 HM_{gas} \quad (4)$$

Where:

$HM_{r,m}$: Input flow rate of each metal contained in raw material, in kg/h.

$HM_{a,f}$: Input flow rate of each metal contained in alternative fuel, in kg/h.

$HM_{f,f}$: Input flow rate of each metal contained in fossil fuel, in kg/h.

$HM_{clinker}$: Output flow rate of each metal contained in clinker, in kg/h.

HM_{gas} : Output flow rate of each metal contained in furnace gas, in kg/h.

D. Measurements of temperature and concentrations of the emitted gases

After starting the SmartAsh incinerator, the temperature and gases concentrations were taken from the stack. A sensor (DELTA 2000 CD-IV device) was used to measure the generated temperature and was placed on top of the incinerator to record the gases resulting from the incineration process as well as their composition. The probe associated with the analyzer detects the concentrations of different gases such as CO, CO₂, NO, NO₂ and SO₂. Table I summarizes the range of concentrations detected by the probe and the sensitivity of the device.

TABLE I: GAS CONCENTRATION RANGES DETECTED BY THE DELTA 2000 DEVICE

Sensor	Detection range	Sensitivity
CO	0 - 4.000 ppm / 10.000 ppm	± 20 ppm or 5 % < 4.000 ppm
CO(*)	0-4% / 10%	± 200 ppm or 5 % read. < 40.000 ppm 10% reading > 40.000 ppm
CO(*)	0 - 4.000 / 20.000 ppm	± 100 ppm or 5 % read. < 4.000 ppm 10 % reading > 4.000 ppm
CO _{2calc}	0 - 20 %	
NO	0-1.000 ppm / 5.000ppm	± 5 ppm or 5 % reading < 1.000 ppm 10% reading > 1.000 ppm
NO ₂	0 - 200 ppm / 1.000 ppm	± 5 ppm or 5 % reading < 200 ppm 10 % reading > 200 ppm
SO ₂	0- 2.000 ppm/ 5.000	± 10 ppm or 5 % reading < 2.000 ppm

ppm 10% reading > 2.000 ppm

III. RESULTS AND DISCUSSION

Increasing the quantity of fuel or changing its composition, by adding solid waste as alternative fuels, lead to the emission of gases, namely nitrogen oxides (NO_x) and sulfur oxides (SO_x). Therefore, it is advisable to control waste ratio and check its composition before the firing process. In fact, the injection of fuels derived from solid waste into the rotary kiln allows their mixture with the raw material. As a result, the heavy metals contained in the waste are combined with those of the raw material during the process of clinker production [1], [4]. The present study highlights the impact of solid waste use on the concentration of the emitted gases and the content of heavy metals throughout the transformation process of raw material into clinker.

A. Impact of the use of solid waste as alternative fuel on the cement production

The problem facing the cement manufacturer was to take into account several parameters when using alternative fuels in cement production. Thus, it was necessary to monitor continuously the content of the pollutants existing in both fuels and raw material. When the limit concentrations stipulated by the directives are exceeded, the dosage of the quantity of the injected fuel must be repeated until an optimal ratio was obtained. This was particularly of great relevance when several types of fuels were used.

In this section, experiments were conducted to estimate the mass balance of heavy metals during a test run using solid recovered fuels. At first, analyses were performed on a representative sample of the waste, then further samples were taken from the raw material (flour) at the kiln entrance, pet coke, and clinker produced by a Moroccan cement plant.

1) Heavy metals in solid fuels

In order to determine the heavy metals contained in the waste, representative samples were analyzed and the results were gathered in Table II.

TABLE II: HEAVY METALS CONCENTRATIONS (PPM) IN SOLID RECOVERED FUELS

FUELS											
Limit values [4]	Maximum values										
	Plastics	Textile	Tetrapack	Blister	Sawdust	Paper	PEBD		PEHD		
Cardboard + Al							Cardboard				
Cd	-	-	-	-	-	-	8.7	-	-	8.7	2
Cr	3	23	32	29	11	84	26	32	84	100	
Pb	36	18	630	95	37	76	50	67	630	200	
Zn	8	209	181	120	110	1710	558	597	1710		
Mg	199	315	384	145	407	732	267	105	732		
Co	-	-	-	91.3	-	104	103	74	104	20	
Ni	7	7	67.9	4.02	39.1	5.9	12	9	67.9	100	
As	-	-	-	-	-	1	-	-	1	15	
Cu	83	21	-	13.5	9	354	4.2	-	354	100	

The results showed that some wastes included elements which concentration was above the limit values, namely textile and blister. Indeed, the measurements exhibited maximum values regarding Cd, Cr, Zn, Mg, Co and Cu for textile, whereas high content of Pb and Ni was detected in the case of blister. In order to dispose of waste used as fuel in cement manufacturing process, it was necessary to consider not only the limit values of the alternative solid fuels but also those of the raw materials, since both were combined during the firing process.

2) Heavy metals in raw materials and cement

Heavy metal concentrations in both raw material and cement were determined and the results were presented in Table III. Examination and analysis of the results obtained showed either a loss or gain of heavy metals during the process of transformation of raw material into cement (clinker). The lost material likely ends up in dust and/or flue gases in the stack. Heavy metals were chemically bound to the organic matrix or concentrated in the mineral fraction. In the course of the organic matrix combustion, heavy metals were released and undergo different processes according to their physico-chemical properties (evaporation, oxidation, etc.). At this point, heavy metal concentrations must be measured in stacks in order to quantify contents of pollutants in dust and flue gases.

From the results listed in Table III, the following points were noticed:

- Arsenic, cadmium and zinc were slightly evaporated. Thus, one can suggest that these elements remain trapped in clinker (98%).
- Copper was partially evaporated (30 to 40%). It can be condensed and re-deposited in the furnace and in the dedusting system.
- Lead and chromium were the most volatile elements. They evaporated during firing and condensed mainly in the stack filters as dust.
- Conversely, in the case of nickel, a material gain was observed during the transformation of raw material into clinker. The noticed augmentation of nickel content was probably due to its presence in the fuel matrices.

TABLE III: HEAVY METALS IN RAW MATERIAL AND CEMENT

Element	Concentration (mg/kg)		Variation %
	Raw material	Cement	
Arsenic	22.1	16.3	26.24
Cadmium	1.47	1.3	11.56
Zinc	57.3	51.2	10.64
Cobalt	0	0	-
Copper	24.15	15.65	35.19
Nickel	24.95	36.5	46.2
Lead	38.05	9.83	74.16
Chromium	150	44.2	70.53

TABLE IV: HEAVY METAL CONCENTRATIONS (PPM) IN THE STACK

Test No	Heavy metals concentrations in mg/Nm ³ à 10% O ₂		Average Concentrations in mg/Nm ³ at 10% O ₂	Sum of average concentrations in mg/Nm ³ at 10% O ₂	Sum of concentrations limit value in mg/Nm ³ at 10% O ₂
	1	2			
Date of test	02/02/2017	02/02/2017			
As	0.0066	0.0036	0.0051	0.1077	0.5
Cd	0.0075	0.0006	0.0041		
Cr	0.0123	0.1058	0.0591		
Co	0.0007	0.0007	0.0007		
Cu	0.0021	0.0021	0.0021		
Mn	0.0021	0.0021	0.0021		
Ni	0.0116	0.0108	0.0112		
Pb	0.0050	0.0314	0.0182		
Zn	0.0075	0.0028	0.0051		

3) Heavy metals in dust and flue gases

Table IV showed the heavy metal concentrations collected at the kiln stack, as well as the referential of heavy metals concentrations. The purpose of presenting these results was to give an idea on the emission gases concentrations typically found in stacks. The results revealed that heavy metals concentrations in the stack remained relatively low and below the standard limits.

4) Mass balance of heavy metals

The average sums of the input and output elements were presented in Table V, along with the calculations of the mass balance of heavy metals. The following remarks could be drawn:

- In the case of arsenic, cadmium, lead, nickel, zinc, chromium and copper, it could be seen that the values of the input and output sums are practically similar. Therefore, it can be suggested that material conservation law was verified.
- For chromium, a loss of material could be noticed, which was likely due to chromium volatilization during the process.

TABLE V: MASS BALANCE OF HEAVY METALS IN THE PROCESS INPUTS AND OUTPUTS (PPM)

Input elements				Output elements			
Raw material (flour)	Pet coke	Waste (SRF)	SUM	Clinker	Stack	SUM	
As	22	2	0	7.7	39	0.005	9
Cd	1.5	0.5	0	0.5	0.6	0.004	0.14
Cr	150	1	57	53.6	42	0.06	9.7
Pb	38	10	56	14.6	77	0.018	17.8
Ni	25	40	21	9.9	46	0.01	10.6
Zn	57	50	670	34	145	0.005	33.5
Co	0	1	1	0.1	9	0.001	2.1
Cu	24	2	56	9.6	49	0.002	11.3

B. Characterization of flue gases

In this section, the interest was directed towards the study of the incineration of non-hazardous solid waste under the effect of air acceleration. For this purpose, the evolution of solid waste temperature and the concentration of the main gases generated from the incineration process were discussed below.

1) Control of flue gas temperature

The flue gas temperature was monitored for the different types of waste samples, namely paper, wood and textile by using the Delta 2000 device. The values were recorded within a 1 min interval. The numerical data are depicted in Fig.4.

In the case of paper waste (Fig.4a), it was noticed that the highest temperature value (about 305 °C) was reached within the first 10 minutes. After 7 minutes, temperature variations were reduced, yield steady values, and then incineration ends. At the beginning, the high temperature increase could be related to a high oxidation of the organic matter, which favored and facilitated the incineration of the sample.

For wood waste (Fig.4b), the Delta 2000 device recorded maximum temperature values around 210 °C. The dense composition of the wood slowed down the flame propagation, and consequently the oxidation process was retarded in comparison to that of paper. Hence, the slow rise in temperature was attributed to a high endothermic degradation of the fuel. After 35 min, a slight temperature rise was detected, which could be related to the oxidation of the carbonized residue. 65 minutes later, the recorded values stabilized around 200 °C.

As to textile waste (Fig.4c), it is noticeable that the maximum temperature value recorded is 270 °C. After 20 minutes of combustion, the temperature drops steadily until it reaches about 80 °C. In fact, the temperature drop is in part caused by the acceleration of the air flow.

Fig.4. Temperature evolution of the combustion of paper (a), wood (b) and textile (c).

2) Composition of incineration gases

In order to discuss accurately the analyses of the emitted gases, it was interesting to recall first the elementary composition of the samples in their raw state. The content of carbon, hydrogen and sulfur contained in the studied samples was illustrated in Table VI.

TABLE VI: ELEMENTAL COMPOSITION OF THE STUDIED WASTE

Sample	% Carbon	% Hydrogen	% Sulfur
Paper	36.7	5.2	0.2
Wood	43.3	6.5	0.1
Synthetic textile	54.6	5.3	0.2

Flue gases values, (CO_2 , CO , NO , NO_x , SO_2), were determined using the Delta 2000 gas analyzer. Measurements were taken within 10 minutes' interval. The evolution of carbon monoxide for the three types of waste were presented separately for a better reading of the values (Fig.5). As to the other emitted gases, the results were shown for each type of waste (Fig.6, Fig.7 and Fig.8).

Fig.5: Evolution of monoxide carbon (CO) concentration in the different samples as a function of time

Fig.6: Evolution of the gases CO_2 , NO , NO_x , SO_2 concentrations in paper waste as a function of time

Fig.7: Evolution of the gases CO_2 , NO , NO_x , SO_2 concentrations in wood waste as a function of time

Fig.8: Evolution of the gases CO₂, NO, NO_x, SO₂ concentrations in textile waste as a function of time.

a) Carbon monoxide and carbon dioxide

During the incineration test, one may notice that CO and CO₂ were the main combustion products and that the quantity of CO₂ emitted depended on the type of fuel. It was noteworthy to mention that wood waste exhibited the highest CO₂ and CO value (maximum concentration of 5% CO₂ and 2800 mg/m³ CO). Moreover, the interesting levels of CO recorded showed that the combustion was not complete at the beginning, especially since the medium was barely starting to be enriched with O₂ required to incinerate the samples. For wood waste, the slow formation of CO₂ could be explained by the oxidation of the carbonized residue resulting from the transformation of CO into CO₂, since the air flow was injected continuously in the course of the combustion. This also justified the temperature increase observed in the temperature evolution diagram (Fig.4b).

As to paper and wood waste, high levels of carbon monoxide are detected if compared to textile waste (rags). This could be justified by the composition of these two fuels (paper and wood) [16], [17]. In fact, they were characterized by a cellulosic composition C₆H₁₁O₅, which facilitated their incineration. Conversely, polyester textile was in the form of acrylic synthetic fiber (C₃H₃N)_n, which required more oxygen according to equations (5 & 6), and hence the reaction tacked longer in comparison to paper and wood.

b) Nitrogen oxides

It can be seen that NO and NO_x concentrations were low compared to carbon oxides. On one hand, maximum NO values were recorded for paper waste (105 mg/m³), which was the main specie of NO_x produced during paper combustion. On the other hand, maximum NO_x values were detected for wood waste (112 mg/m³). The diagrams showed that NO was not the only gas emitted among NO_x gases. Moreover, it is noticeable that NO_x formation in the case of wood was slower than that of paper. This could be attributed to the slow kinetics of NO_x formation in comparison to the oxidation rates of hydrocarbon species [18], which explained the reason why NO_x generation tacked about 30 min to occur.

Furthermore, the elemental analysis revealed that nitrogen exists in low concentrations in the waste samples. Thus, NO_x gases were produced mainly from the nitrogen in air and partially from nitrogen contained in small quantities in the fuel. The thermal decomposition of the nitrogen compounds (aniline, pyridine, pyrroles, etc.) contained in the fuel generates species and radicals such as HCN, NH₃, N-, CN- and NH-, which can be converted to NO_x.

c) Sulfur oxides

During the incineration of solid waste samples, sulfur dioxide was formed in small quantities due to the low initial concentration of sulfur in the samples. In comparison to paper and textile wastes, wood waste had the highest maximum value of about 54 mg/m³.

IV. CONCLUSION

Through this study on non-hazardous solid waste incineration and its use in cement manufacturing process, several interesting results were highlighted and could be listed as follows:

- Heavy metals such as lead and chromium volatilize during firing process, while arsenic, cadmium and zinc were trapped in clinker.
- For gaseous emissions, heavy metals concentrations in the stack remained relatively low and below the standard. This proved that the disposal of waste in cement plants remained one of the least polluting processes regarding atmospheric emissions.
- The combustion regime was characterized by low reaction temperatures, which promoted endothermic degradation and high CO production.
- Temperature rise and continuous injection of air promoted the transformation of CO into CO₂ (reduction of CO efficiency).
- NO production was controlled by O₂ concentration: greater air flow induced important NO concentration.
- Wood waste exhibited high concentrations of gaseous pollutants such as CO, CO₂ and NO_x if compared to textile and cardboard wastes.
- Paper incineration emitted high concentrations of CO, NO and NO_x.
- The concentrations of gases emitted by textile waste remained the lowest in comparison to those of wood and paper wastes.

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