Research Article

Comparative study on the energy efficiency of the calcination and of the hot mixture of phosphate: Application of Bouchane phosphate (Morocco)

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A b s t r a c t

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Experimental tests of a new heat treatment protocol by hot mixture the phosphate of Bouchane (Morocco) which aims at optimizing spending energy for calcination has already been presented in our previous work. These studies allow us to conclude that the procedure of calcinations followed by cooling by 33% of phosphate can have a significant impact on energy saving with increased chemical reactivity while preserving the environment. In this paper, we try to evaluate and assess the energy efficiency of our new protocol of hot mixture by drawing an energy balance sheet of calcination and of hot mixture in order to establish the feasibility of our heat treatment protocol. This approach should lead in principle to a reduction in energy costs up to 33% and an increase in productivity and chemical reactivity during the attack by sulfuric acid.

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1. Introduction

Environmental quality and energy efficiency play an increasingly important role in energy processes, whether in new construction or in renovation. The issue is related to the reduction of energy consumption in the sector of energy processes is first of all ecological: it comes to reducing emissions of greenhouse gases so as to protect the planet against the climate change. Thus, it requires an effective technology transfer process with low impact on the greenhouse effect. The improvement of energy efficiency reduces the greenhouse gas emissions by reducing emissions that comes from the

measures which aims at achieving greater energy efficiency, taking into account the different scenarios **[1]**. This efficiency is directly proportional to the reduction of energy. It is a way for the public service to act on the equilibrium between supply and the demand [2]. The tests profitability of energy efficiency measures based on the idea that energy efficiency measure must allow the reduction of energy consumption in various social and economic sectors [3 - 5].

In general, the energy savings from 10 to 30% can be obtained by techniques of process integration. Thus, organizations can, in order to prolong their policy and existing energy choices, opt for process optimization, energetic analysis and other methods based on thermodynamics [6]. The optimization process, taking into account the current requirements in quality, security and environment and sustainable development objectives, represents a relatively virgin area for scientists and industries. It pushed us to have time to any of the new technologies and innovations.

The choice of energy source, the process and method of heat recovery is critical to the design of the furnace, and these are also among the main factors that influence the environmental performance and the energy efficiency of manufacturing the process [3 - 5].

More specifically, the thermal treatment of phosphate, which represent approximately 95.7% of mining production in Morocco, requires huge amounts of energy for the removal of unwanted materials, which in addition to the fuels are the two main sources of polluting the environment. This is associated with problems of energy loss. For this reason, the recycling and / or the recovery of energy is essential for energy recovery. In recent decades, many studies have been carried out and developed, in several rich countries in phosphates in order to develop treatment and recovery methods for energetically less costly phosphate. However, the results are insignificant [6 - 9] because of the complexity of these granular materials. This complexity is due to the diversity of physical properties and chemical and crystallographic characteristics where impurities are responsible [7]. It is on the basis of this diversity that we can determine the appropriate type of treatment. The presence or not of impurities modifies significantly the kinetics of heat treatment.

In the majority of works [7 –14], the removal of the organic matter and carbonates and heavy metal stabilization is carried out through the calcination process of phosphate that is based on the fluidized bed. This type of process offers many advantages in particular; it encourages the chemical reactions and ensures efficient heat transport [8, 13, 14]**.** However, the problem of energy consumption remains a serious problem in all calcination process that are usually done at a high temperature followed by cooling to ambient temperature. For this reason, it is not a desirable operation in all sedimentary phosphate ores due to the contribution and significant energetic losses and high cost of the energy [8], [13 - 16], as well as, the issuance of greenhouse gas emissions whose impact on the environment is well established. It was also observed that the calcination decreases the performance of the physical properties of phosphate, in particular, the solubility and reactivity of the ore [13, 14]. One is thus

confronted with the problem of optimization and innovation of a thermal process, suitable and somewhat flexible.

The experimental protocol of hot mixture which was previously proposed [18, 19] allows by passing these problems by reusing the energy associated with the calcination for the treatment of another quantity of phosphate. Indeed, we were able to demonstrate that the addition of a quantity of 33% of the gross from the same source at the beginning of the cooling cycle phosphate calcined at 800 °C provides a phosphate concentrate better with the added growth of the chemical reactivity and solubility [18, 19]. This advantage will lead to limit the rate of sedimentation of the phosphate pulp and reagents during the attack.

The purpose of this paper is to evaluate and assess the energy efficiency of our new heat treatment protocol for hot mixture [18, 19]. Thus, we examine and quantify the heat energy required to perform each step of the process. We draw up, accordingly, an energy balance sheet of the calcination and hot mixture.

2. Description of the calcination process and of our hot mixture protocol

2. 1. Description of the calcination process

The majority of calcination of phosphate operations occurs according to the process ″DORR-OLIVER″ which is based on the fluidization [17]. From the point view of energy, the calcination occurs generally in three stages:

a) Production and distribution of the energy: the combustion of organic matter and fuel in the calciner are the main sources of this energy.

b) Recovering energy: this is the operation of cooling the product.

c) Reusing the recovered energy: it is the process of drying and preheating.

The figure 1 shows the descriptive scheme of the calcination process and its energy distribution.
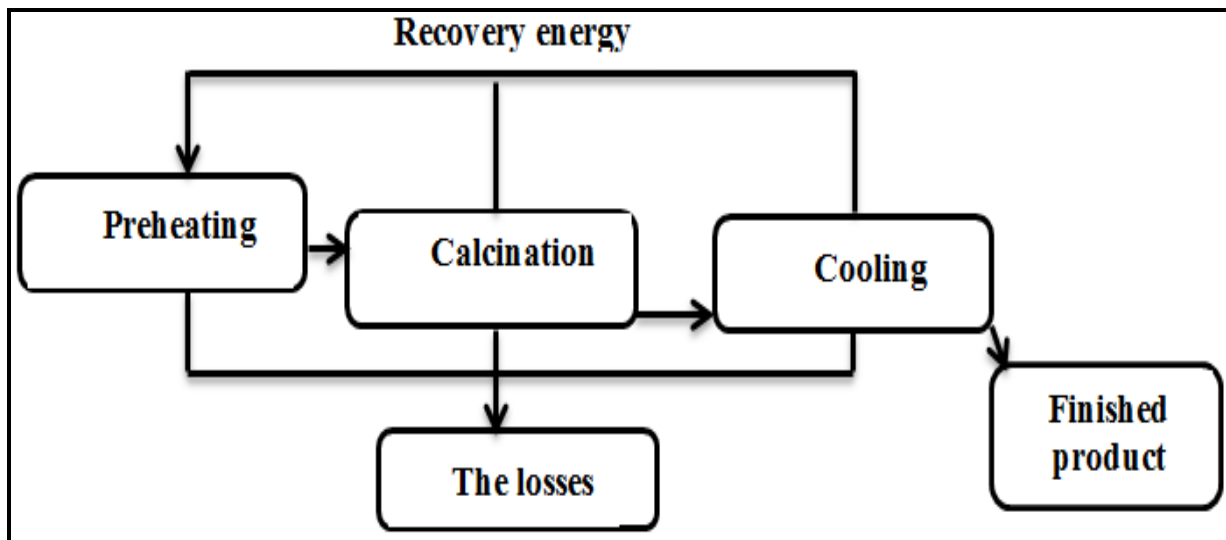
Fig. 1: Schematic of the calcination process

2. 1. Description of the hot mixture protocol

The heat treatment operation by hot mixture is proposed in the same manner as the calcination (Figure. 1) by adding a phosphate appoint between the phases of the calcination and cooling. Thus, the chillers that are fluidized beds in the process **"DORR-OLIVER"** will be substituted by a rotary mixer in the treatment by a hot mixture where the mode of heat transfer is direct between the calcined phosphate (the hot) and raw phosphate (the cold). The figure. 2 is a descriptive diagram of the protocol of hot mixture.
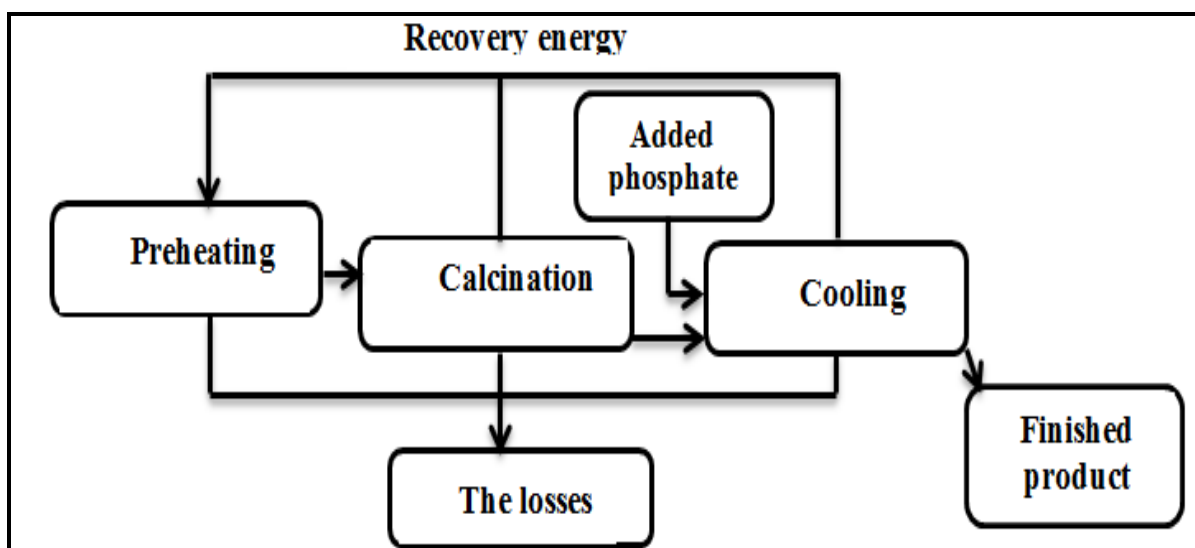

Fig. 2: Schematic representation of the processing by hot mixture

3. Energy calculation of heat treatment of phosphate

Thermodynamics is a concept and a basic tool for energy applications and for some environmental applications. In our case of the thermal treatment of phosphate, the balance sheet of the energy is based on the determination of heat that was at stake along all unit operations of the process. The mineralogical characterization and preliminary determination of specific heats, that are related to the different constituent phases of phosphate, is an essential step in the calculation.

3. 1. The mineralogical phases that constitute the phosphate of the study area

The phosphate of Bouchane is a carbonate fluorapatite [18, 19]. The mass percentages of mineralogical characteristics of dry phosphate are summarized in Table. 1. The main constituents of phosphate ores are the apatites which are associated, with clays and silica minerals, carbonates, sulphates and organic matter.

Table 1: Phases of mineralogical characteristics of Bouchane phosphate and molar masses

Phase	Mass percentage (%)	Molar mass (g/mol)
$Ca_{10}(PO_4)_6F_2$	90.66	1008.6
$CaCO_3$	3.28	100.09
$CaMg(CO_3)_2$	3.13	184.40
C_{Org}	1.48	12
SiO_2	1.45	60.08

3. 2. Specific heats of the phases that constitute the studied phosphate

The specific heat at constant pressure (Cp) is one of the most important greatness of thermodynamic, because it allows access to variations in enthalpy as a function of temperature for a medium studied. The data of the specific heat of phosphates are not available in the literature. To overcome these shortcomings, we propose in the following an estimation of this greatness by quantum methods. These are based on theoretical calculations that take into account the electronic structure of the studied systems and the resolution of the Schrödinger equation. The implementation of these calculations is used in the form of computer softwares. In this study, we used the Gauss View Software 5.0.8, to estimate the specific heats of the different phases of phosphate. The results of this simulation are shown in Figure 3.
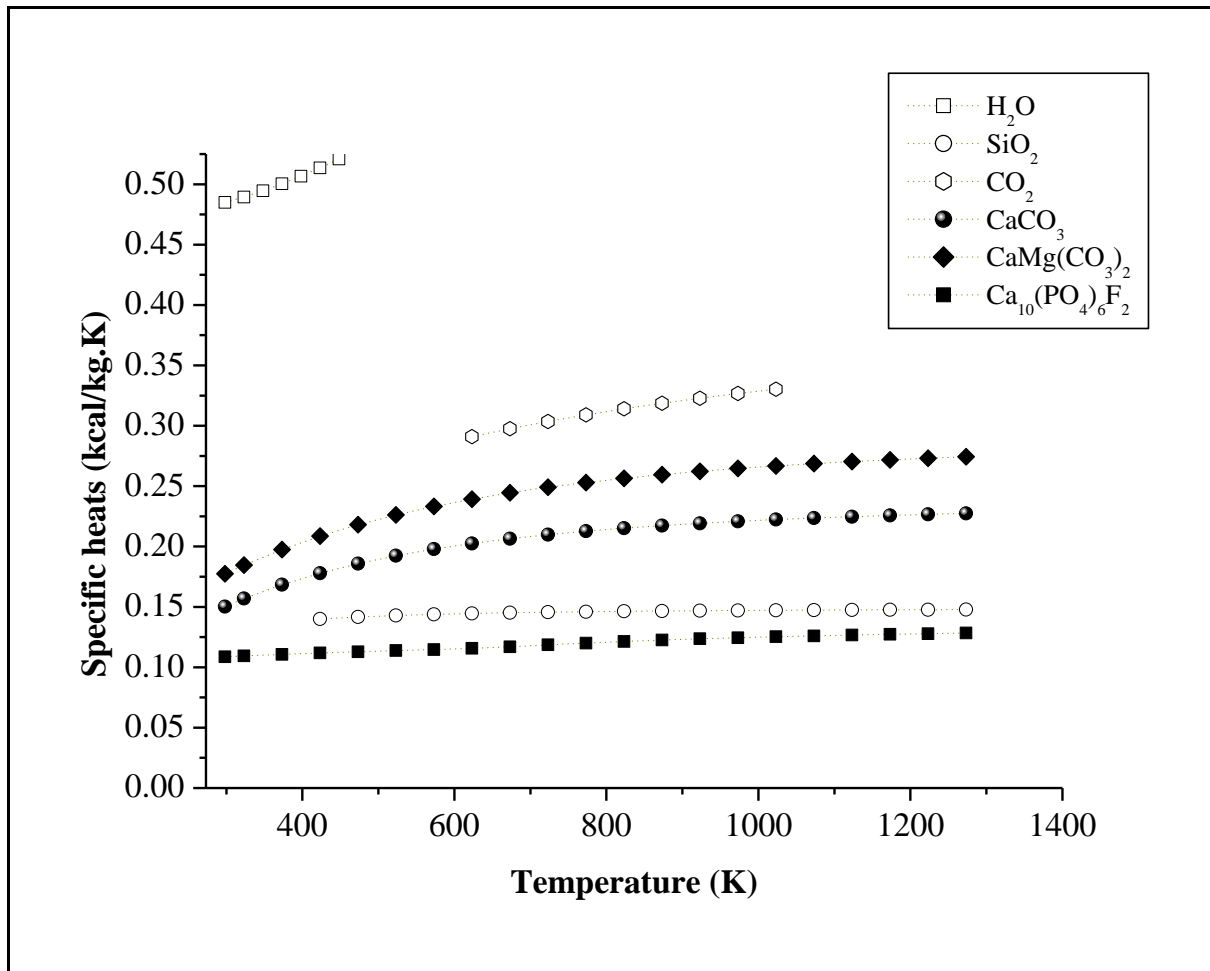
Fig. 3: Specific heats of different phases of Bouchane phosphate

The variations of specific heats of the different phases of phosphate are slightly slower between 278.15 and 1073.15 K. Therefore, the specific heat of phosphate varies significantly in the same way in this temperature range as in that of the fluorapatite. It is deduced by using the following formula:

$$C_p = \frac{\sum_{i=1}^{n} P_i C_{pi}}{100}$$

(1)

- C_p : The specific heat of phosphate;

- C_{pi} : The specific heat of phase ″i″ ;

- P_i : The percentage by mass of phase ″i″ ;

- n : Number of phases.

Table 2: Expressions of specific heats of water, phosphate and carbon dioxide in different temperature intervals

	Specific heats (kcal/kg.K)	Temperature intervals (K)
Water	$Cp_w = 0.394 + 2.83 \times 10^{-4}T$	298.15 to 373.15
	$Cp_v = 0.394 + 2.83 \times 10^{-4}T$	373.15 to 403.15
Phosphate	$Cp_p = 0.126 + 0.33 \times 10^{-4}T$	298.15 to 403.15
	$Cp_p = 0.095 + 0.259 \times 10^{-4}T$	403.15 to 673.15
	$Cp_p = 0.103 + 0.209 \times 10^{-4}T$	673.15 to 1073.15
Carbon dioxide	$Cp_{CO2} = 0.23 + 9.752 \times 10^{-4}T$	673.15 to 1073.15

4. Energy analysis of the calcination

In this paragraph we will try to calculate the different energies highlighted in the energy balance sheet calculations while neglecting the calorific losses by radiation.

4. 1. Heat needed for drying and preheating phosphate

The drying operation and preheating consists of preheating the crude phosphate to a temperature of about 400.15 K in order to vaporize the water content (generally 97% of the water is evaporated). This is realized thanks to calories brought by gases from the fire losses during the phase of calcination and by a part of the energy recovered from the cooling phase.

The energy required for preheating the phosphate (Q_1) is a sum of the following energies.

a) The energy of overheating moisture (q_H) contained in the raw phosphate at the entrance of the preheater, of the ambient temperature until around 403.15 K: This sensible heat is given by the following formula :

$$q_H = m_H \times \int_{298.15}^{373.15} Cp_w dT + m_H \times L_v + m_H \int_{373.15}^{403.15} Cp_v dT \tag{2}$$

With   the first term is the energy of heating the humidity;

the second term represents the latent heat of vaporization of water ;

the third term is the energy of the superheat of water vapor.

and is the specific latent heat of vaporization of water at 373.15 K.

One can also write q_H as a function of mass of humidity as follows:

$$q_H = 15.12 \times m_H + 540 \times m_H + 36.67 \times m_H \tag{3}$$

Or :

$$q_H = 591.79 \times m_H \tag{4}$$

b) The energy to carry the ore to the superheat temperature (403.15 K) is given by the following relationship:

$$q_p = m_P \int_{298.15}^{403.15} Cp_p \, dT \tag{5}$$

With m_p is the mass of the sec phosphate. One obtains:

$$q_p = 14.43 \times m_p \tag{6}$$

Consequently, the energy (Q_1) required to preheat the raw phosphate at a temperature of 403.15 K is the sum of the total energy of the superheat of humidity and the energy to carry the ore preheating temperature. This energy can be written as follows:

$$Q_1 = q_H + q_p \tag{7}$$

We found:

$$Q_1 = 591.79 \times m_H + 14.43 \times m_P \tag{8}$$

It appears that the preheating energy depends on the humidity contained in the phosphate. Therefore, the minimizations of this energy requires a reduction in the rate of humidity of phosphate entering the preheater. This is possible, for example by performing a pre-drying of phosphate in the open air by exploiting the dry climate of the zones of the heat treatment.

4. 2. Energy for the calcination of phosphate

This energy (Q_2) involves one part for carrying the ore from 403.15 to 673.15 K, then a second part driven by fire losses and finally the energy for grilling and decarbonating ore that is operating in the temperature range from 673.15 to 1073.15 K. Thus, we write its expression as follows:

$$Q_2 = m_P \int_{403.15}^{673.15} Cp_P \, dT + m_{FL} \int_{673.15}^{1073.15} Cp_{CO_2} dT + m_P \int_{673.15}^{1073.15} Cp_P dT \tag{9}$$

Or :

$$Q_2 = 75.12 \times m_P + 126.06 \times m_{FL} \tag{10}$$

We considered that the fire losses are triggered by 673.15 K. In the expression (9) we assumed for simplicity that only carbon dioxide is released during calcination.

5. The total energy of the calcinations

The energy (Q) for calcining the raw phosphate is the sum of the different energies previously determined:

$$Q = Q_1 + Q_2 \tag{11}$$

It can be written as follows:

$$Q = 591.79 \times m_H + 89.55 \times m_P + 126.06 \times m_{FL} \tag{12}$$

We recall that the energy for drying and preheating is recovered from calcination (all fire losses) and in partly from cooling. Therefore, only the energy Q_2 is theoretically required during calcination and must supplied by the combustion of fossil fuel. In other words, Q_2 must supply the calciner in normal functioning. This energy depends on the respective masses of dry phosphate and of fire losses. By not taking into account the energies associated with endothermic and exothermic phenomena since for the phosphate of Bouchane, these enthalpies are relatively the same magnitude [18, 19].

The fules commonly used are diesel or fuel. Take for example the case of a process powered by fuel. This fuel has a lower calorific value (LCV) to be worth 9650 kcal/kg. By not taking into account the combustion performance, the minimum mass of fuel (m_F) to be used is given by the following expression :

$$m_F = \frac{Q_2}{PCI} = \frac{75.12 \times m_P + 126.06 \times m_{FL}}{PCI} \tag{13}$$

6. The energy lost in the cooling phase

Our objective is to recover the energy generated by the cooling phase for use in the treatment of another quantity of phosphate. Theoretically, after the calcination the energy lost (Q_L) during the cooling phase of phosphate is estimated by:

$$Q_L = Q_2 - Q_1 \tag{14}$$

It can be written as a function of the phosphate masses, fire losses and the humidity as follows:

$$Q_L = 60.69 \times m_P + 126.06 \times m_{FL} - 591.79 \times m_H \tag{15}$$

It is found that the energy lost in the phase of cooling increases with increasing fire losses and decreased rate of humidity. Therefore, the amount of treated phosphate will be important.

7. Calculation of the energy efficiency of the calcinations

The energy consumed in the calcination process is calculated by formulas (10) and (14). We deduce that:

$$Q_C = Q_1 \tag{16}$$

Or:

$$Q_C = 14.43 \times m_P + 591.79 \times m_H \tag{17}$$

Energy efficiency (η_C) of the calcination is defined as the ratio of energy necessary to conduct the process and the energy supplied to the calcinator.

$$\eta_C = \frac{Q_C}{Q_2} \tag{18}$$

According to formulas (10) and (17), energy efficiency can be expressed as functions of the respective masses of dry phosphate, humidity and the fire losses as follows:

$$\eta_C = \frac{14.43 \times m_P + 591.79 \times m_H}{75.12 \times m_P + 126.06 \times m_{FL}} \tag{19}$$

It is concluded that the energy efficiency of heat treatment operations depends on the humidity rate, the carbonates that are associated with endothermic reactions and the organic matter that is associated with exothermic reactions.

8. Calculation of the energy efficiency of the treatment by hot mixture

The energy required to process a third of the added raw phosphate is calculated as follows:

$$Q_{1/3} = \frac{Q}{3} \tag{20}$$

It can be written in terms of the respective masses of dry phosphate, the fire losses and humidity. Thereby:

$$Q_{1/3} = 197.26 \times m_H + 29.85 \times m_P + 42.02 \times m_{FL} \tag{21}$$

The energy supplied to the calcinator is the same as in the case of hot mixture without calcination. Thus, the energy used in the protocol of hot mixture is derived by the following equation:

$$Q_{HM} = Q_2 + \frac{1}{3}Q_2 - \frac{Q}{3} - Q_1 = Q_2 - \frac{4}{3}Q_1 \tag{22}$$

It can also be expressed by the following relationship:

$$Q_{HM} = 55.88 \times m_P + 126.06 \times m_{FL} - 789.05 \times m_H \tag{23}$$

The energy consumed in the hot mixture is calculated by formulas (10) and (23). We find after simplification:

$$Q_{CHM} = Q_2 - Q_{HM} = 19.24 \times m_P + 789.05 \times m_H \tag{24}$$

We deduce the energy efficiency of the treatment of hot mixture by the following equation:

$$\eta_{HM} = \frac{Q_{cmc}}{Q_2} = \frac{19.24 \times m_P + 789.05 \times m_H}{75.12 \times m_P + 126.06 \times m_{FL}} \tag{25}$$

Energy efficiency depends on the mass of phosphate, humidity rate and the rate of conversion of degradables matters. For a given phosphate, only the humidity rate can vary. To examine the effect of this parameter on the efficiencies η_C and η_{HM}, we consider the example of a mass $m_0 = 1000$ kg of raw phosphate. The Table 3 summarizes the results found.

Table 3: Comparison of efficiencies and masses of finished products of calcination and hot mixture
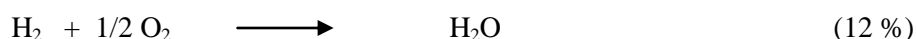
H (%)	m_P(kg)	m_H(kg)	m_{FL}(kg)	η_C (%)	H_{HM} (%)	m_C(kg)	m_{HM}(kg)
8	920	80	73.6	77.3	100	846.4	1128.5
7	930	70	74.4	69.2	93.3	855.6	1140.8
5	950	50	76.0	53.5	73.6	874.0	1165.3
2	980	20	78.4	31.1	44.2	901.6	1202.1

According to the Table 3, we notice that the efficiency in the case of hot mixture is more interesting than that in the case of the calcination. This means that as long as humidity is reduced the

quantity of the treated phosphate is larger. In our case, the treatment by hot mixture, the energy efficiency is interesting for a humidity of H = 8% and an addition of 33%. For humidity H <8%, the amount of added phosphate which must be treated is more than a third. So for humidity H> 8% the quantity of phosphate is treated less than a third. This amount can be increased by adding fuel surcharge.

9. Estimation of CO_2 attenuation

Energy is the blood of the economy and a key to the environment protection, the calcination of phosphates generates significant carbon dioxide emissions due not only to the energy consumption but also to do wire netting of the organic matter and decarbonation of the carbonates. The energy of the calcination proceeds from the combustion of the main fuel where the combustion reactions are:

$$C \ + \ O_2 \longrightarrow CO_2 \qquad (84\ \%)$$

$$H_2 \ + \ 1/2\ O_2 \longrightarrow H_2O \qquad (12\ \%)$$

$$S \ + \ O_2 \longrightarrow SO_2 \qquad (4\ \%)$$

The different CO_2 emissions produced during the processing of phosphate by calcination (PC) and by hot mixture (PHM), are calculated and shown in Table 4.

Table 4: Comparison between the emissions from the calcination and the hot mixture

H(%)	m_p(kg)	m_f(kg)	CO_{2f} (kg)		CO_{2p}(kg)		CO_2 total (kg)		$kgCO_2/kgpp$	
			PC	PHM	PC	PHM	PC	PHM	PC	PHM
8	920	8.1	6.8	6.8	73.6	98.1	80.4	104.9	0.095	0.093
7	930	8.2	6.9	6.9	74.4	99.2	81.3	106.1	0.095	0.093
5	950	8.4	7.1	7.1	76.0	101.3	83.1	108.4	0.095	0.093
2	980	8.7	7.3	7.3	78.4	104.5	85.7	111.8	0.095	0.093

It is found that the emissions in the heat treatment protocol of hot mixture are lower than in the case of calcination. Thus, the CO_2 emission factor decreases by 2 kg of CO_2 per ton of the produced phosphate (2 $kgCO_2/10^3$ kgpp). Also, there is a decrease by one third of the fuel which is used in processing the amount of the added raw phosphate.

10. Conclusion

The comparative study on the energy efficiency of the calcination and hot mixture of the phosphate of Bouchane judged and evaluated the socioeconomic and environmental importance of our protocol hot mixture. It confirms to us the increase in energy efficiency of an interesting value. This value is dependent on the humidity rate. Thus, we observed a decrease by 2 kg of CO_2 emissions per tone of the produced phosphate with an increase in the quantity of processed phosphate.

It will be understood that the hot mixture with proportions of 75% of calcined and 25% of raw may have an impact on energy consumption (up to 33%) and the reduction of the emission of greenhouse gases, especially CO_2, more than one third (1/3). In addition, we found through the analyzes carried out [18, 19] that the velocity of cooling of hot mixture phosphate provokes an increase in the specific surface and decrease in the density; and consequently increase the reactivity. These structural variations make the product of a hot mixture more reactive during the manufacturing of phosphoric acid by attacking the product obtained by hot mixture by the sulfuric acid.

Nomenclature

Latin letters		
C_{Org}	Organic carbon	----
C_p	Specific heat capacity of phosphate	$kcal.kg^{-1}.K^{-1}$
C_{pi}	Heat capacity of the phase "i"	$kcal.kg^{-1}.K^{-1}$
IGCE	International group of climat experts	----
L_W	Mass latent heat of water at 373.15 K	kcal/Kg
M_i	Molar mass of constituent "i"	Kg / mol
m_0	Mass of raw phosphate	kg
m_H	Mass the humidity of phosphate	kg
m_P	Mass of dry phosphate	kg
M_{FL}	Mass of fire losses	kg
m_c	Mass of the finished product from the calcination	Kg
M_{HM}	Mass of the finished product from the hot mixture	Kg
m_f	Mass of fuel	Kg
n	Number of phases	----
P_i	Weight Percent of phase "i"	%
NCV	Net calorific value	Kcal/Kg

PC	Phosphate treated by calcination	----
PHM	Phosphate treated by hot mixture	----
Q	Total energy of the calcination phosphate	kcal
Q_1	Energy preheating of crude phosphate	kcal
Q_2	Energy for the calcination of phosphate	kcal
Q_L	Energy lost during the cooling phase	kcal
q_H	Energy overheating of the humidity contained in the raw phosphate	kcal
Q_p	Energy to carry the ore superheat temperature	kcal
Q_C	Energy consumed in the process of calcination	kcal
Q_{HM}	Energy used in the protocol of hot mixture	kcal
Q_{CHM}	Energy consumed in the hot mixture	kcal
$Q_{1/3}$	Energy required to treat a third of the added raw phosphate	kcal
Greek letters		
η_C	Energy efficiency of the calcination	%
η_{HM}	Energy efficiency of the hot mixture	%
Indices		
p: phosphate v: vapor w : water CO_2: carbon dioxide f : fuel		

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