

RESEARCH ARTICLE

Material transfer during the combustion of Tarfaya oil shale or Jerada coal (Morocco), simulation and diffusion laws

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Manuscript Info

Abstract

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The study of the oxidation or combustion of oil shale or coal is important because it is the basis of the exploitation process to obtain the energy to be converted This exploitation involves several physical phenomena such as heat transfer, matter transfer and others. The laws of matter transfer have been adopted The respective combustion of two national energy materials (Morocco), namely the Z4 layer of oil shale from the Tarfaya deposit and the coal from Jerada, was the subject of this study in an isothermal regime. First, we were interested in mentioning the average temperatures remarkable for this combustion which turned out to be slightly different, then we started a study of the combustion of parallelepipeds with variable dimensions of the two previous materials and we presented a simulated theoretical model by comparing it with the experimental results. This simulated model allowed us to discuss the reactivity of the two energetic materials. Combustion was followed in an isothermal regime using a muffle furnace from 400°C to 800°C for fine-grained samples (< 0.5 mm) and for grains. The noticeable loss of mass for oil shale at 500°C, and for coal at 600°C in the first instance. These remarkable temperatures were used to study the effect of parallelepiped dimensions, which have constant height (z = 2 mm) and square side surfaces (x = y = 2, 4, 6, 8, 10 mm). Several results and interpretations were noted when comparing the combustion of coal and oil shale.

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Introduction:-

The study of the diffusion of matter during the combustion of coal or oil shales has interested us, when we speak of diffusion we usually refer to Fick's laws (Adolf. Fick: 1855). The oxidation of oil shale and combustion is important because it is the basis of the "in situ" exploitation process. Reference is made to the work done by the authors (R.G.Mallon et al: 1975), (A.L.Tyler/ 1975) and (Huang: 1977) on the thermal behavior in air of shale blocks where Mallon et al observed abnormally high velocities near the center of a shale block. The mechanism known so far is not satisfactory to explain this behaviour. Indeed, the transport of oxygen by diffusion through the surface of the block towards the interior is weak enough to cause an abrupt rise in temperature. The same is true for the authors (Tihen et al: 1968), who calculated the thermal diffusibility and showed the opposite phenomenon, i.e. that the temperature rise at the surface is faster than in the interior.

Corresponding Author:-Abdeljabbar. Attaoui Address:-DepartmentofChemistry,FacultyofSciencesBenm''sikCasablanca,UniversityHassanIIMorocco. In the course of this work, we will vary the dimensions of the material to be studied, whether it is oil shale or coal. In powder or grain form, both materials will undergo combustion at different temperatures. The appropriate mechanism for this kind of dimensioning of the two materials is the volume contraction mechanism (**C.Lee and H.Y.Sohn: 1986**). Two temperatures were found to be remarkable for working the parallelepiped shape (x=y variable from 2mm to 10mm with constant z equal to 2mm), these temperatures are 500°C for oil shale and 600°C for coal.

1/ Bibliographical study

Authors (A.Iven and J.Jacobson: 1980) have studied the oxidation at decomposition temperatures of shale under an atmosphere consisting of N_2 , O_2 , CO_2 , CO and H_2O . Their mathematical model is based on the contraction of the sample volume. Furthermore, it is the diffusibility of oxygen in the shale that delimits the rate of the reaction. According to the same authors, when studying the combustion of shale in a horizontal reactor and under the same conditions, the percentage of oil production is less than 50% compared to the percentage in the Fisher test. The combustion kinetics of five Australian oil shale samples was studied at 900°C using a fluidized bed reactor. Gas analyses are determined on a continuous basis (B.G. Charlton: 1988). All the degraded oil shale show that the combustion reaction follows the volume contraction model and the model is controlled by diffusion. The diffusibility of the gas is controlled by diffusion and is related to the diffusion through the spent shale. The kinetic parameters and activation energies are low compared to carbon combustion, and the combustion temperatures are low compared to those of American shale. The same author (B.G. Charlton: 1988) has shown that the volume contraction model applies to the Naggorin shale, and that this mechanism is attributed to the reaction of highly concentrated carbon.

By similarity the author (**B.K.Mazymard et al: 1957**)oxidised coals in air and the authors (W.E. Robinson et al: 1963) oxidised Colorado shale at a temperature of 170°C for 14 weeks in air as well. They noted that the H/C ratio decreased from 1.59 to 0.63 while the O/C ratio increased from 0.1 to 0.46. The authors (**J.H Levy and KI Stuart: 1984**) studying the combustion of kerogen show that it evolves in two stages which are indicated by the presence of two peaks in DTG. The first peak is due to the reaction of the aliphatic compounds, while for the second peak they suggest that it is the reaction of the aromatic chains that is predominant. Similarly, the author (**P.C. Uden: 1980**) and the author (**C.M. Earnest: 1983**), when oxidizing shale in air, noted the presence of two stages which vary in position and intensity in the thermogravimetric diagram, depending on the composition of the organic matter and in some cases that of the mineral matter. The authors (**C.Lee and H.Y.Sohn: 1986**) studied the oxidation kinetics of Colorado shale in air using the technique of thermogravimetric analysis in a linear and continuous temperature rise. Their kinetic data are analyzed at low temperatures, conditions under which they claim that the oxidation reaction occurs at the surface of the solid (kerogen) at temperatures where the kerogen does not decompose. Furthermore, the oxidation rate is first order with respect to the partial pressure of oxygen, the calculated activation energy is: (11.0 ± 2.3) kJ/mol and the frequency factor is (6.8 ± 2.5) 10^{-6} m (kPa s)⁻¹.

Consider a sphere of initial radii ro reacting with oxygen, the rate of heterogeneous surface kinetics is written as follows:

-I/S dm/dt = K ρP_{02}^n According to I.C Lee and H.Y. Sohn m = quantity of kerogen in kg. S = area of the interfacial reaction in m². ρ = bulk density in kg/m³ K = rate constant in m.atm-n s⁻¹.

After transformation the model of the volume contraction in dynamic regime ($\theta = dT/dt = heating rate$) is. $d\alpha/dT = 3k_0 P_{02}^n (1-\alpha)^{2/3} exp (-E/RT) /\theta r_0$

According to this equation, the order with respect to the chemical reaction is 2/3 and the order with respect to the oxygen partial pressure is n. This model reflects the total decomposition of a sphere and without residue.

Oil shale from the Rundle deposits in Queensland, Australia (John S. Killingley and D. Glen Callaghan: 1988) were decomposed and then underwent combustion. The characteristics of the release of sulphurous and nitrogenous

gases were studied during combustion in a small reactor connected to a mass spectrometer. Hydrogen cyanide is the dominant nitrogenous gas released below 500°C, while at higher temperatures nitric oxide is dominant. The release temperature of sulphur dioxide from pyrite oxidation is related to its size, which explains the release of sulphur dioxide from shale sulphides at temperatures below 500°C during combustion. The amount of sulphur dioxide evolved from shale decomposed during isothermal combustion between 600 and 880°C is independent of temperature. Natural samples of silicate minerals found in Devonian shale (kaolinite, illite, quartz) have also been tested (**S. D. Carter. D. N. Taulbee and T. L. Robi: 1993**). The rate of carbon uptake decreases with increasing carbon deposition on the solids. Shale coke activities are classified as: shale undergoing combustion, gaseous shale; shale undergoing pyrolysis. The effect of coke temperature on carbon deposition is relatively small, which has important implications for recycled oil shale solids.

In oil shale processes where the pyrolysis zone is heated by solid recirculation, the adsorption of oil vapor and subsequent coking and cracking on these solids can lead to the reduction of significant oil yields.

It has been shown that the type of solid heat carrier can have a significant effect on the extent of oil loss (**T.T.Coburn:1988 and P.Udaja,G.J.Duffy and M.D.Chensee:1990**). The authors' interest in this section focuses on the coke trends of the materials used for heat transfer in the Kentort II process. The Kentort II process includes zones of pyrolysis, gasification and combustion between which the processed shale is recycled to transfer heat.

For the combustion of coal we refer to work done at different pyrolysis temperatures (Shengping Ma, J. O. Hill and S. Heng J: 1989), these authors studied by the thermal analysis methods TG, DTG and DTA. The non-coking of coal produces a char during the pyrolysis process. Coal char is mainly used in the manufacture of acetylene in the plastics industry. The char properties depend on the conditions of the pyrolysis, through which it is produced.

Thermal analysis provides a quick method of determining the oxidation process of char. The DTA curve reflects the magnitude of the enthalpy change during combustion, and the TG and DTG profiles can be used to derive the corresponding kinetic parameters.

Several studies have been reported on the reactivity of coal char using the TG isotherm technique (**M.Rashid Khan: 1987, R.Suhu et al 1988).** The results obtained show that at low temperature chars show higher reactivity for coal or char at high temperature, and that the chemical adsorption capacity of oxygen is an important reactivity parameter.

The authors (**M.M.Patel et al: 1988**) studied the rate of combustion of lignite char by isothermal methods. The activation energy obtained in the controlled chemical frequency region is 120 Kj/mol compared to the isothermal analysis, the non-isothermal analysis has some advantages. A mass loss curve equivalent to a large number of isothermal mass loss curves and only one is required. Victorian lignite coal and char are very reactive to oxygen. It is very difficult to use isothermal techniques to study the kinetics of oxidation, as the sample reacts immediately after exposure to oxygen and the combustion reaction is therefore exothermic and the temperature of the sample is higher than that of the furnace.

Concerning the environment we note that these two materials have differences in the release of carbon dioxide (CO2) during combustion:

 $\begin{array}{ccc} Coal & \xrightarrow{Combustion} & CO_2 \ + \ residue \end{array}$

Whereas oil shale which are formed by hydrocarbons (kerogen) release CO₂ and H₂O:

Combustion

Oil shale \rightarrow CO₂ + H₂O + residue This reduces the release of the greenhouse gas

2/ Experimental study2.1: Raw material.2.1.1: Oil shale.

Oil shale are two generic terms indicating fine-grained sedimentary rocks containing mineral matter and organic matter called kerogen, which by pyrolysis can yield an oil that resembles oil and gas.

Oil shale can be burned directly as a quality fuel for power generation in the form of electricity, and can be used as a feedstock in the chemical and building materials industries.

2.1.2: Coal.

Coal is a black solid combustible rock, mainly consisting of carbon, hydrogen, oxygen and to a lesser extent nitrogen, sulphur and minerals. Coal is a very heterogeneous material in terms of its physical and chemical properties, and its composition differs from one deposit to another or even from one extraction to another. This heterogeneity is due to the diversity of the initial plants (trees, ferns, algae) and the conditions of their decomposition and fermentation (time, temperature, pressure, oxygen content).

2.2: Isothermal combustion of oil shale (Z4) and coal at different temperatures

Our work consists of comparing the combustion of oil shale and coal, for which we used Tarfaya oil shale from the Z4 layer and Jerada coal, in the first part of this work, the raw rock from the oil shale and coal were first crushed by a manual crusher, part of it was ground very finely to obtain a pulverized powder and another part was passed through a sieve to obtain grains with a size of about4 mm.

1g of each sample was weighed and placed in a crucible, then both samples (powder and grains) were placed in a muffle furnace where they underwent combustion at 400°C in an isothermal regime, this combustion was carried out for 30 min with an access of air to the interior of the furnace every 10 min, then the experiment was repeated for 500, 600, 700 and 800°C, and finally the samples were weighed again after combustion.



Fig 1:- Muffle furnance.



Fig 2:- Balance.



Fig 3:-Coal and oil shale samples.

The results obtained are shown in the table below:

Table 1:- Percentage loss of mass of the powdered samples for the different temperatures.

Tuble II Teleentt	ige loss of mass of the powdered samples for the a	interent temperatures.
	Coal	Oil shale

Pulverizing	Initial mass	Remaining mass	Percentage loss	Initial mass	Remaining mass	Percentage loss
400 °C	0,997 g	0,960 g	3,71 %	0,960 g	0,785 g	21,81 %
500 °C	0,999 g	0,890 g	10,91 %	0,890 g	0,719 g	28,31 %
600 °C	1,003 g	0,457 g	54,43 %	0,457 g	0,685 g	31,84 %
700 °C	1,003 g	0,359 g	64,20 %	0,359 g	0,574 g	42,66 %
800 °C	1,004 g	0,261 g	74 %	0,261 g	0,500 g	50,15 %

Table 2:- Percentage of mass loss of the samples in the form of grains for the different temperatures.

	Coal			Oil shale		
	Initial mass	Remaining	Percentage	Initial	Remaining	Percentage
Grains		mass	loss	mass	mass	loss
400 °C	0,997 g	0,938 g	5,92 %	1,003 g	0,783 g	21,93 %
500 °C	1,003 g	0,900 g	10,27 %	0,995 g	0,680 g	31,66 %
600 °C	0,997 g	0,588 g	41,02 %	1,001 g	0,673 g	32,77 %
700 °C	1,005 g	0,394 g	60,79 %	0,996 g	0,572 g	42,57 %
800 °C	0,998 g	0,187 g	81,26 %	1,006 g	0,319 g	68,29 %

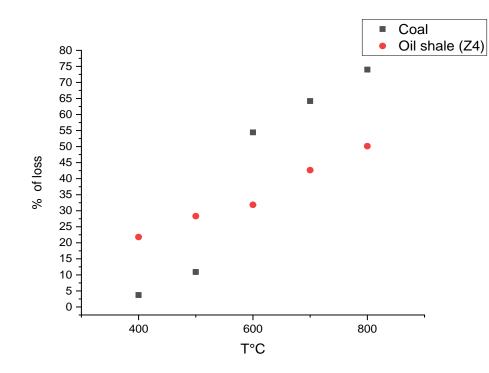


Fig 4:- Percentage mass loss of oil shale and powdered coal as a function of temperature.

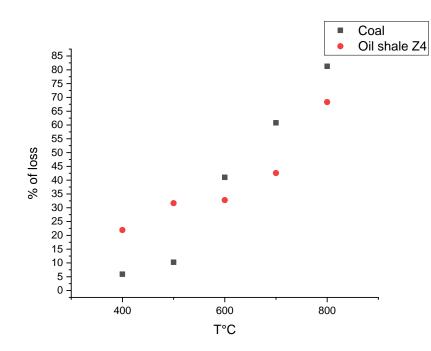


Fig 5:- Percentage mass loss of oil shale and coal in grain form as a function of temperature.

The remarkable temperatures for the evolution of mass loss are 500, 700 and 800°C where we have significant losses for oil shale. This is similar to what happens in pyrolysis, where up to 500°C we have the decomposition of organic matter and then a pseudo-palate from which we have the decomposition of carbonates.

As regards coal, we notice a significant loss up to the temperature of 600°C, then from this temperature onwards we have a loss that aligns with the temperature.

The same remarkable temperatures are repeated for the samples in pulverized form or in high granulometry.

So in the chosen temperature range we have a higher loss for oil shale than for coal up to 500°C, then after 600°C it is coal that decomposes more under air than oil shale.

2.3: Effects of sample dimensions during combustion of oil shale (Z4) and coal

For the second part of the work, the same experiments are repeated, except that this time the focus is on specific temperatures, i.e. 500° C for oil shale and 600° C for coal. The samples to be studied are of different sizes, first pulverized, then in parallelepiped form (with a height of z = 2 mm, and a square surface with sides x or y = 2, 4, 6, 8 and 10 mm).

The results obtained are shown in the table below:

	Coal (600°C)	Coal (600°C)			ale (500°C)		
	Initial mass	Remaining	Percentage	Initial	Remaining	Percentage	
		mass	loss	mass	mass	loss	
Pulverising	1,003 g	0,457 g	54,43 %	1,003 g	0,719 g	28,31 %	
2 10 ⁻³ m	1,008 g	0,636 g	36,90 %	1,026 g	0,757 g	26,22 %	
4 10 ⁻³ m	0,995 g	0,646 g	35,07 %	1,003 g	0,725 g	25,73 %	
6 10 ⁻³ m	0,997 g	0,689 g	30,89 %	1,004 g	0,753 g	25 %	
8 10 ⁻³ m	1,003 g	0,705 g	29,71 %	1,015 g	0,759 g	25,22 %	
10 10 ⁻³ m	1,006 g	0,720 g	28,43 %	0,996 g	0,755 g	24,19 %	

Table 3:- Percentage mass loss of coal at 600°C and oil shale at 500°C for different dimensions.

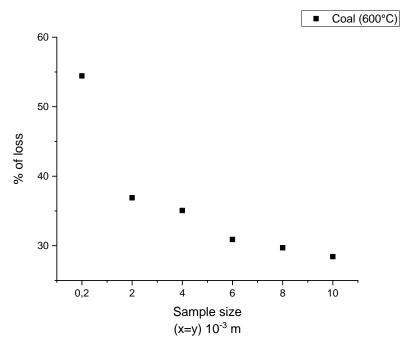


Fig 6:- Percentage mass loss of coal at 600°C as a function of sample size in m.

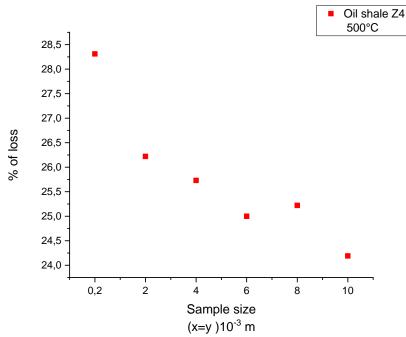


Fig 7:- Percentage mass loss of Z4 oil shale at 500°C as a function of sample size in m.

2.4: Discussion:-

The diffusion of matter by analogy with that of heat (Fick's law and Fourier's law) is managed by the following empirical law:

DQ = -D (dC / dn) dSdt

with Q: material transfer

n: space vector (three dimensions)

S: equipotential surfaces transmitting matter in one direction

The material flow rate dQ/dt = -D (dC / dn) dS

and the material flow Q = - D (dC / dn)

 $Q = -D \operatorname{grad} C$

This is Fick's first law

For a permanent regime without material source, which is our case, in fact we were working in an isothermal regime where all the samples are thermally exposed causing the decomposition of the material during an identical time. Also, the samples are weighed at the same mass and placed in the same muffle furnace, which imposes the conditions of the regime without material source, so we have the Laplacian $\Delta C = 0$

The shape of our samples is parallelepipeds of identical height (z = 2 mm) and variable x = y square area of 2, 4, 6, 8 and 10 mm

We will adopt the Laplacian for one x direction only, hence:

dC /dx

C being the concentration for what concerns the Fick equation, concerning our conditions we consider the percentage of loss for a pulverizing product and parallelepipeds of fixed height (z = 2 mm) and variable square surface x = y of 2, 4, 6, 8 and 10 mm. The mass loss after plotting (Figures: 6 and 7) for coal and for oil shale varies at first sight by falling exponentially as a function of x.

3/ Simulation and experimental results for this reaction.

After integration the adopted model is as follows:

$$P(x) = P (pulverizing) / e^{1000 x \log K}$$

with x in m

By simulation with the experimental results the exponential constant k is:

K = 1.1278 for coal

K = 1.0248 for oil shale

The experimental results and those adapted by the simulated theoretical model are shown in the following tables:

• For coal :

	P _{practice}	P _{simuleted}
Pulverising	54,43 %	54,429 %
2 10 ⁻³ m	36,90 %	42,79%
4 10 ⁻³ m	35,07 %	33,64 %
6 10 ⁻³ m	30,89 %	26,45 %
8 10 ⁻³ m	29,71 %	20,81 %
10 10 ⁻³ m	28,43 %	16,35 %

• For oil shale :

	P _{practice}	P _{simuleted}
Pulverising	28,31 %	28,18 %
2 10 ⁻³ m	26,22 %	27,07 %
4 10 ⁻³ m	25,73 %	25,90 %
6 10 ⁻³ m	25 %	24,77 %
8 10 ⁻³ m	25,22 %	23,70 %
10 10 ⁻³ m	24,19 %	22,67 %

The following figures 8 and 9 represent this simulation and the experimental results of the combustion of the two national products. The exponential character is observed in a large way for the case of coal that the shale.

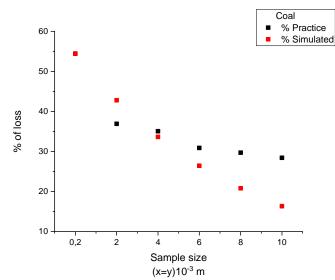


Fig 8:- Curve of practical and simulated percentage mass loss during coal combustion at 600°C as a function of sample size.

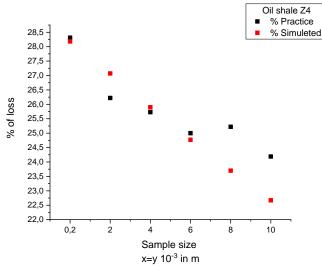


Fig 9:- Curve of practical and simulated percentage mass loss during combustion of oil shale at 500°C as a function of sample size.

4/ Determination of the diffusion coefficient and validity of Fick's first law.

By applying the empirical law of matter transfer for the test x=10mm (P (10mm)) we could determine the diffusion coefficient at the considered temperatures:

dQ = -D (dC/dx) dSdt

By observing the curves, both the experimental and the simulated ones, we notice a rather clear linearity between:

x = 2 (sample size) and x = 10 for coal

x = 2 (sample size) and x = 10 for oil shale

The Laplacian will be adopted for a single x direction, hence : dC / dxHow do we determine D the diffusion coefficient?

dQ = -D (dC/dx) dSdt

For oil shale

dQ = total diffuse material for the test x=10mm =10 10^{-3} m=0.2419 g / 6 parallelepipeds For 1g we have 6 parallelepipeds dC = total material / unit volume = $(1g/10.10.2 \ 10^{-9})/6$ parallelepipeds dS = $(10.10.2 + 10.2.4)10^{-6}$ dx varies from 0 mm to 10 mm so the - is cancelled out 0.2419 g / 6 =- D ((0.996/10.10.2 \ 10^{-9}. 6). ((10.10.2 + 10.2.4)10^{-6})/0-10.10^{-3}).1800

The number of parallelepipeds simplifies on both sides, which means that the coefficient depends only on the mass taken from the sample. $DC = 9.6410^{-10} \text{ m}^2 \text{s}^{-1}$

d Q = total diffuse material for the test x=2mm =0.2622 g/n parallelepipeds np = number of parallelepipeds dC = total material / unit volume = $(1.026/2.2.2 \ 10^{-9})$ / n parallelepipeds dS = $(2.2.2 + 10.2.4)10^{-6}$ dx varies from 0 mm to 2 mm so the - is cancelled DC=0.95 $10^{-10} \text{ m}^2\text{s}^{-1}$

Dimension	Diffusion coefficient of oil shale	Diffusion coefficient of coal
$x = y = 2 \ 10^{-3} m$	$0,95 \ 10^{-10} \ \mathrm{m}^2 \mathrm{s}^{-1}$	$1,36 \ 10^{-10} \ \mathrm{m^2 s^{-1}}$
$x = y = 4 \ 10^{-3} m$	$2,85 \ 10^{-10} \ \mathrm{m^2 s^{-1}}$	$3,92 \ 10^{-10} \ \mathrm{m^2 s^{-1}}$
$x=y=6 \ 10^{-3}m$	$4,9 \ 10^{-10} \ \mathrm{m}^2 \mathrm{s}^{-1}$	$6,1 \ 10^{-10} \ \mathrm{m}^2 \mathrm{s}^{-1}$
$x=y=8 \ 10^{-3}m$	$7,54 \ 10^{-10} \ \mathrm{m}^2 \mathrm{s}^{-1}$	$8,6\ 10^{-10}\ \mathrm{m}^2\mathrm{s}^{-1}$
$x=y=10 \ 10^{-3}m$	$9,64 \ 10^{-10} \ \mathrm{m^2 s^{-1}}$	$11,21 \ 10^{-10} \ \mathrm{m}^2 \mathrm{s}^{-1}$

We will plot the diffusion coefficients as a function of x, a single direction in Figure 10

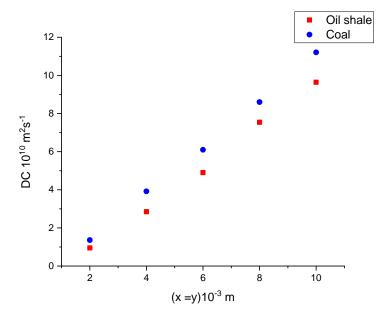


Fig 10:- Diffusion coefficients of the two compounds oil shale and coal.

Dimension of parallelepiped x=y=2mm, z=2mm ; x=y=4mm, z=2mm ; x=y=6mm, z=2mm ; x=y=8mm, z=2mm and x=y=10mm, z=2mm

We observe two well-aligned straight lines from which we can calculate their respective slopes We do the calculation for x=4 and x=6 we find the line and its slope for Oil shale: $CD=1,025 \times 10^{-7} - 1, 25 \times 10^{-10}$ Coal: $CD=1, 09 \times 10^{-7} - 0, 44 \times 10^{-10}$

We take x in meter

Conclusion:-

As the study of the combustion reaction is very important as it is the reaction responsible for the production of thermal energy, this work was initiated for two national energy raw materials, namely Tarfaya oil shale (Z4) and Jerada coal.

The combustion was followed in isothermal regime thanks to a muffle furnace from 400°C to 800°C for samples of fine granulometry (powder) (g < 0.5 mm) and for grains.

In the chosen temperature range we have a higher loss for oil shale than for coal up to 500°C, then after 600°C it is the coal that decomposes more under air than the oil shale.

The noticeable loss of mass for oil shale at 500°C, and for coal at 600°C in the first instance. These remarkable temperatures were used to study the effect of parallelepiped dimensions, which have a constant height (z = 2 mm) and square side surfaces (x = 2, 4, 6, 8, 10 mm).

The first result is that the mass loss decreases in a high way when x increases and also compared to a sample of fine grain size (< 0.5 mm). Looking at the curve P = f(x) for shale, two bounded domains of x are presented from P pulverizing to P(x = 2 mm) and from P(x = 2 mm) to P(x = 10 mm).

Fick's law of matter transfer has been verified where we have linearity of the diffused matter as a function of the parameter x between x = 2 mm and x = 6 mm. Similarly for the coal between x=2 mm and x=10 mm.

The variation of the experimental results led us to consider a theoretical simulated model that could interpret in a detailed way this variable size combustion of the sample. The model is presented as follows:

$$\begin{split} P(x) &= Pulverizing \ / \ e^{1000 \ x \ \log k} \\ \text{with:} \quad k &= 1.1278 \ \text{for coal} \\ k &= 1.0248 \ \text{for oil shale} \end{split}$$

As the percentage of loss is inversely proportional to this exponential constant k, the latter has informed us about the reactivity of these two materials, which is more sensitive for oil shale than for coal.

As for the diffusion coefficients, we have calculated them by adopting Fick's law for the permanent regime, and these coefficients, as we have noted, line up exactly according to the following two lines:

Oil shale: $CD= 1,025 \times 10^{-7} - 1,25 \times 10^{-10}$ Coal: $CD= 1,09 \times 10^{-7} - 0,44 \times 10^{-10}$

with x in meter.

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