

Variation of Feed Chemical Composition and Its Effect on Clinker Formation–Simulation Process

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Abstract— The raw materials for Portland cement production are the mixture (as fine powder in the 'Dry process') of minerals containing calcium oxide, silicon oxide, aluminum oxide, ferric oxide, and magnesium oxide. The raw materials are usually quarried from local rocks which are found in some places have already practically the desired composition and in other places require addition of the clay and limestone, as well as iron ore, or recycled materials such as CKD]. The homogeneity of feed chemical composition has an important relationship to fuel consumption, kiln operation, clinker formation and cement performance.

Cement quality is typically assessed by its compressive strength development in mortar and concrete. The basis for this property is a well-burned clinker with consistent chemical composition and free lime. The main reason for the clinker free lime to change in a situation with stable kiln operation is variation in the chemical composition of the kiln feed. This variation in chemical composition is related to raw mix control and the homogenization process. To ensure a constant quality of the product and maintain a stable and continuous operation of the kiln, the attention must be paid to storage and homogenization of raw materials and kiln feed. This article focuses to study the role of raw mix composition control and the homogenization process. The study have been done by computer simulation (Fortran 90) to make calculations to estimate quantities of the raw materials (limestone, clay, iron ore,...) required to prepare kiln feed. These calculations have been done based on knowing composition of raw materials, these field data obtained from Al-Mergheb cement plant. Due to variations in the kiln feed chemical compositions that affect its burnability and eventually the fuel consumption, so the simulation done to give a suitable composition of kiln feed to allow soft burning inside kiln.

A simulation of the mixing raw materials process and estimate composition of kiln feed, as well as formed clinker were done successful through fitting of the results obtained with field data of one of the plants operating in Libya.

Index Terms—Clinker, Limestone, Cement Quality, Portland Cement.

I. INTRODUCTION

Portland cement (often referred to as OPC, from Ordinary Portland Cement) is the most common type of cement in general use around the world because it is a basic ingredient of concrete, mortar, stucco and most non-specialty grout. It is

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a fine powder produced by grinding Portland cement clinker. Portland cement clinker is a hydraulic material which shall consist of at least two-thirds by mass of calcium silicates ($3CaO.SiO_2$ and $2CaO.SiO_2$), the remainder consisting of aluminium- and iron-containing clinker phases and other compounds.

There are three fundamental stages in the production of Portland cement [1]:

- Preparation of the raw mixture
- Production of the clinker
- Preparation of the cement

The raw materials for Portland cement production are the mixture (as fine powder in the 'Dry process') of minerals containing calcium oxide, silicon oxide, aluminum oxide, ferric oxide, and magnesium oxide. The raw materials are usually quarried from local rocks, which in some places is already practically the desired composition and in other places require addition of the clay and limestone, as well as iron ore, bauxite or recycled materials [2, 3].

The homogeneity of feed chemical composition has an important relationship to fuel consumption, kiln operation, clinker formation and cement performance.

Cement quality is typically assessed by its compressive strength development in mortar and concrete. The basis for this property is well-burned clinker with consistent chemical composition and free lime. There are only two reasons for the clinker free lime to change in a situation with stable kiln operation: variation in the chemical composition of the kiln feed or variations in its fineness. Variations in fineness depend on possible changes in raw materials or in operation of the raw mill. Variation in chemical composition is related to raw mix control and the homogenization process. To ensure a constant quality of the product and maintain stable and continuous operation of the kiln, attention must be paid to storage and homogenization of raw materials and kiln feed. This article is focused to study the role of raw mix composition control and the homogenization process. The study has been done by computer simulation (Fortran 90) to make calculations to estimate quantities of the raw materials (limestone, clay, iron ore,...) required to preparation feed to kiln. These calculations done based on knowing composition of raw materials, these field data are obtained from Al-Mergheb cement plant, (*Alahlia Cement Company at Libya*). Variations in the kiln feed chemical composition affect its burnability and eventually the fuel consumption. So the calculations done to give a good raw feed composition to the kiln to allow soft burning inside kiln.

II. CLINKER FORMATION AND BURNING PROCESS IN ROTARY KILN

A. Clinker Formation

Portland cement clinker mainly consists of CaO , SiO_2 , Al_2O_3 and Fe_2O_3 ; are accounting for more than 95%. The

minor components in total less than 3% are MgO , TiO_2 , P_2O_5 and alkalis. In clinker, they are not present in individual oxide, but exist as compounds formed by two or more oxides. The mineral phases are very fine, usually 30- 60 μm and consist mainly of alite, belite, calcium aluminate and aluminoferrite.

The process of clinker formation is described in Fig.1. The transformation concludes with the primary clinker phases [4]:

- Alite (C_3S) is the most important constituent, 50-70% in normal Portland cement clinkers. It is tricalcium silicate (Ca_3SiO_5) modified in composition and crystal structure by ionic substitutions.
- Belite (C_2S) constitutes 15-30% of normal Portland cement clinkers. It is dicalcium silicate (Ca_2SiO_4) modified by ionic substitutions and normally presents wholly and largely as the P-polymorph.
- Aluminate (C_3A) constitutes 5-10% of most normal Portland cement clinkers. It is Tricalcium aluminate ($Ca_3Al_2O_6$), substantially modified in composition and sometimes also in structure by ionic substitutions.
- Ferrite (C_4AF) makes up 5-15% of normal Portland cement clinkers. It is a tetracalcium aluminoferrite (Ca_4AlFeO_5), substantially modified in composition by variation in Al/Fe ratio and ionic substitution.

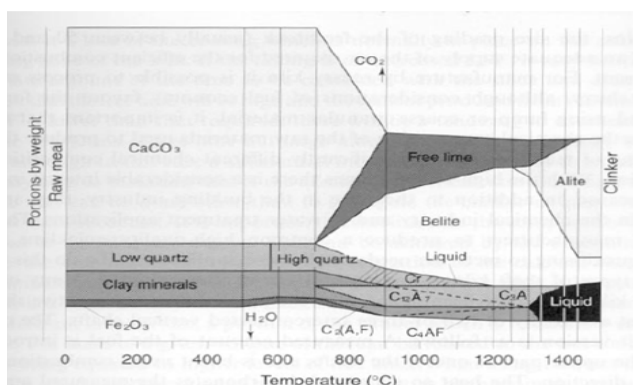
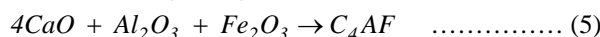


Fig.1 A schematic view of the clinker formation reactions [4]

Reactions taking place in rotary kiln system are illustrated in Fig.1. On the left-hand is the feedstock comprising, in this case, calcite ($CaCO_3$), quartz (SiO_2), clay minerals ($SiO_2-Al_2O_3-H_2O$) and iron oxide (Fe_2O_3). Up to a temperature of about 700°C, activation through the removal of water and crystal structure changes take place. Within the temperature range 700-900°C, decarbonation of the calcium carbonate occurs, together with the initial combination of the alumina, ferric oxide and of activated silica with lime. From 900 to 1200°C, Belite forms. Above 1250°C and more particularly above 1300°C, the liquid phase appears and this promotes the reaction between Belite and free lime to form Alite. During the cooling stage on the right-hand side of the diagram, the molten phase goes to a glass or, if cooling is slow, the C_3A crystallizes out and in extreme cases the Alite dissolves back into the liquid phase and reappears as secondary Belite. Alkali sulfates condense out as a separate phase during the cooling process but these are not shown in Fig.1.

Assumed chemical reactions occur throughout the kiln [4]:



B. Burning Process

In modern systems of cement manufacture, the retention time of the material in the kiln is 30 to 40 minutes, of which the major part is in the burning zone. The temperature of the material increases rapidly from 850°C to 1250°C to 1300°C, at which temperature clinker melt is formed. In the burning zone, the chemical and physical changes of the material take place simultaneously; it is important with respect to the kinetics of clinkerization reactions and agglomeration processes. The exact temperature of melt formation depends on the chemical composition of the feed.

Under certain conditions, melt may be formed at temperatures lower than 1250°C. For instance, quartz grains in the kiln feed will give rise to melt at 1200°C or lower. The surrounding CaO will partly agglomerate and form crystals of Belite, which can be identified in the clinker as Belite nests. Also, the alkalis, sulfates, and chlorides present in the kiln system will form liquid phases at lower temperatures than the proper clinker liquid. However, the largest amount of clinker melt is formed within a narrow temperature interval and over a short distance in the rotary kiln. The coating formed on the brick lining reflects this. In the upper transition zone or the entrance to the burning zone, the soft, relatively thin coating changes to a dark, clinker like, hard, thick coating within half a meter.

So, the amount of the clinker minerals formed depending on the time and temperature treatment of the mix, and overall chemical composition of the kiln feed [4].

If the pyroprocessing time is too short or the temperature too low, combination of the raw material components may be less complete and some free unreacted lime will be present. Insufficient control of the raw mixture and its blending will cause large variations in the chemical composition of the kiln feed. If the kiln is operated at constant material residence time and temperature, that will cause variations in clinker composition, including free lime. This is important because the free lime is usually used as the process parameter to indicate how well the clinker is burned. When unintended variation in kiln feed composition causes large variation in free lime, operators may make incorrect changes to kiln operation, assuming changes are needed when they are not [1].

III. VARIATION EFFECTS OF KILN FEED CHEMICAL COMPOSITION

A. Chemical Composition Variation of Kiln Feed

Variations in the kiln feed chemical composition affect its burnability and eventually the fuel consumption. This relationship has been described elsewhere [2].

Table.1 show the chemical composition of raw materials that used in Al-Mergheb cement plant (one of Alahlia cement company, Khoms city, Libya) where they find it very normal for operation, so require proportions of mix may be obtained easily then fed to kiln to form a clinker.

Table.1 Chemical composition of raw materials used to preparing kiln feed mix [5]

Comp.	Limestone	Marl	Iron ore	Clay	CKD
SiO ₂	8.34	27.98	8.56	53.75	9.55
Al ₂ O ₃	2.74	10.87	6.79	14.4	2.58
Fe ₂ O ₃	0.94	3.08	68.69	4.99	2.37
CaO	47.62	30.12	5.97	8.71	53.27
MgO	0.97	1.95	0.481	1.8	1.1
SO ₃	0	0.7	0.901	0.31	0.301
K ₂ O	0.39	0.2	0.151	1.99	1.08
Na ₂ O	0.04	0.33	0.177	0.62	0.0865
LOI	38.54	24.68	5	13.37	29
Rest	0.42	0.09	3.28	0.06	0.6625
Total	100	100	100	100	100

Chemical compositions of kiln feed that product as result of mixing processes also for the clinker formed throughout the kiln are shown in Table.2, in which the kiln operating at ideal case (normal operating state).

Table.2 Chemical composition of kiln feed mix and clinker formed (field data of ideal operation) [5]

Component	Kiln feed	Clinker formed
SiO ₂	14.43	22.22282
Al ₂ O ₃	3.9	5.878449
Fe ₂ O ₃	2.27	3.450678
CaO	42.61	65.12717
MgO	1.09	1.656632
SO ₃	0.1	0.152684
K ₂ O	0.9	1.374154
Na ₂ O	0.09	0.137415
LOI	34.61	--
Total	100	100
CaCO ₃ content	76.09	--

For the same raw materials shown in Table.1, Table 3 show chemical composition of the final product out of mixing process that fed to kiln (kiln feed), this mixture is hard burning without clinker formation in which the kiln operating is nonideal. Many problems associate with hard burning of process occurs (as discussed previously). As shown in Table 3 found that chemical composition of this kiln feed mixture are not homogeneous as required in terms of the amount of calcium carbonate content is higher than that required for optimum operating (it is 79-79.05%), while for optimum operating the chemical composition of kiln feed must be ranged (76-76.5%), this range give soft burning of kiln feed and leading to clinker formation [5]. Also, amount of fuel used for burning will be in economic range.

This problem occurs in some plants, especially the old-made due to either a faulty balance of raw materials. This would lead to an imbalance in the composition of the raw material fed to kiln, or due to technical problems of monitoring systems of chemical analysis for raw materials, as well as the mix material fed to kiln.

B. Effect on Fuel Consumption, Clinker and Cement Performance

Insufficient mix control or blending will result in larger variations than anticipated in chemical analysis. The result is variations in clinker free lime. The operator may be obliged to increase the burning zone temperature to achieve the desired free lime level by keeping the kiln on the hot side, the maximum clinker free lime is brought to the average value and the spikes are eliminated (*Response: Burn Harder*) [1].

Table.3 Chemical composition of kiln feed mix (field data of nonideal operation) [5]

Component	Kiln feed Case (1)	Kiln feed Case (2)	Kiln feed Case (3)
SiO ₂	11.601	11.61	11.616
Al ₂ O ₃	2.522	2.517	2.52
Fe ₂ O ₃	2.0526	2.056	2.0515
CaO	44.241	44.267	44.268
MgO	1.422	1.413	1.405
SO ₃	0.217	0.222	0.219
K ₂ O	0.784	0.708	0.784
Na ₂ O	0.042	0.088	0.043
LOI	37.2789	37.2897	37.281
Total	100.1605	100.2589	100.2937
CaCO ₃ content	79	79.048	79.05

As a result of this harder burning, fuel consumption increases. The fuel penalty for burning to an average of 0.8% free lime because of large variability instead of an average of 1% can easily be on the order of 4% [2]. When the kiln is operated on the hot side, alkalis and sulfate become more volatile. This, in turn, might increase the possibility for build-ups in the cooler parts of the kiln system. In severe cases, controlling the kiln may become difficult because of surges of the material through the kiln. Hard burning tends to cause low clinker porosity, large crystals of Alite, and often contributes to generation of dust instead of good, nodular clinker [6,7]. It also slows down the cooling process, both because the maximum temperature is higher, and because the low-porosity clinker is more difficult to cool [1].

These effects all can result in cement with reduced strength potential and increased water demand. Reduced clinker porosity can make the clinker harder to grind, increasing finish mill power consumption or reducing mill production. Clinker temperatures exiting the cooler may increase, further increasing fuel consumption and presenting handling problems. The high-temperature conditions may lead to color variations, reductions in clinker alkali and sulfate level, and increases in water demand attributable to increased levels of Aluminate. Variations in clinker alkali and sulfate will affect concrete setting time, and result in strength variations. Periods with decreased clinker alkali content will result in a decrease in early strength and increase in later-age strength; the opposite can occur during periods when the clinker alkali content increases. With such variability, fresh concrete often develops admixture incompatibility and changes in its rheological behavior [1].

IV. CEMENT MODULUS

For a long time cement was manufactured on the basis of practical experience collected from the process of production. When comparing chemical analyses of Portland cement (Feed raw materials and/or clinker) it was found that certain relations exist between the percentage of lime on the one hand and the combination of silica, alumina and iron oxide on the other. These modulus are [8];

A. Hydraulic Modulus(HM)

The hydraulic modulus is generally limited by the values (1.7-2.3), which has the following form;

$$HM = \frac{CaO}{SiO_2 + Al_2O_3 + Fe_2O_3} \dots\dots\dots (6)$$

It was found that with an increasing *HM*, more heat is required for clinker burning; the strengths, especially the initial strengths set up and also the heat of hydration rises; and simultaneously the resistance to chemical attack decreases. Generally cements with *HM* lesser than 1.7 showed mostly insufficient strength; cements with *HM* greater than 2.3 had poor stability of volume.

B. Silica Ratio (SR)

It is the proportion of SiO_2 to the total of Al_2O_3 and Fe_2O_3 , given as following;

$$SR = \frac{SiO_2}{Al_2O_3 + Fe_2O_3} \dots\dots\dots (7)$$

Increasing silica ratio impairs the burnability of the clinker, by reducing liquid phase content and tendency toward formation of coating in the kiln. An increasing silica ratio causes a slow setting and hardening of the cement. With decreasing silica ratio the content of liquid phase increases, this improves the burnability of the clinker and the formation of coating in the kiln. Generally the silica ratio runs between (1.9-3.2), low values for silica ratios can be accepted as low down to 1.5.

C. Alumina Ratio (AR)

It is characterizing the cement by the proportion of alumina to iron oxide, given as following;

$$AR = \frac{Al_2O_3}{Fe_2O_3} \dots\dots\dots (8)$$

Values of alumina ratio are in the range from 1.5 to 2.5. The *AR* determines the composition of liquid phase in the clinker, when its lower than 1.5 both oxides are present in their molecular ratios and therefore only tetracalcium aluminoferrite can be formed in the clinker; consequently, the clinker cannot contain tricalcium aluminate. This is the case called Ferrari-cement which is characterized by low heat of hydration, slow setting and low shrinking. A high alumina ratio together with a low silica ratio results among other things, in a fast setting of the cement; this requires the addition of a higher gypsum rate to control the setting time.

D. Lime Saturation Factor

To attain complete lime saturation in the clinker the total silica must be combined as C_3S , all iron oxide must combine with the equivalent amount of alumina to C_4AF , and the remaining alumina must combine to C_3A . This formula expressed as following for *MgO* lower than 2% [9,10];

$$LSF = \frac{100(CaO + 0.75MgO)}{2.85SiO_2 + 1.18Al_2O_3 + 0.65Fe_2O_3} \dots\dots (9)$$

For technical purposes good values ranged between (80-95).

V. RAW MATERIALS AND COMPOSITION CALCULATIONS OF RAW MIX

A. Mixing Process of Raw Materials

Materials of natural origin as well as industrial products can be used for the production of cement. Starting materials for this purpose are mineral compounds containing the main components of cement: lime, silica, alumina and iron oxide. Seldom are these components founding the needed proportion in only one raw material. Therefore it is usually necessary to select a measured mixture of a high lime component (lime component) with a component which is lower in lime containing however more silica, alumina and iron oxide (clay component) [8].

Fig.2 show the simplified process of mix preparing as required specification to give a soft burning before fed to kiln.

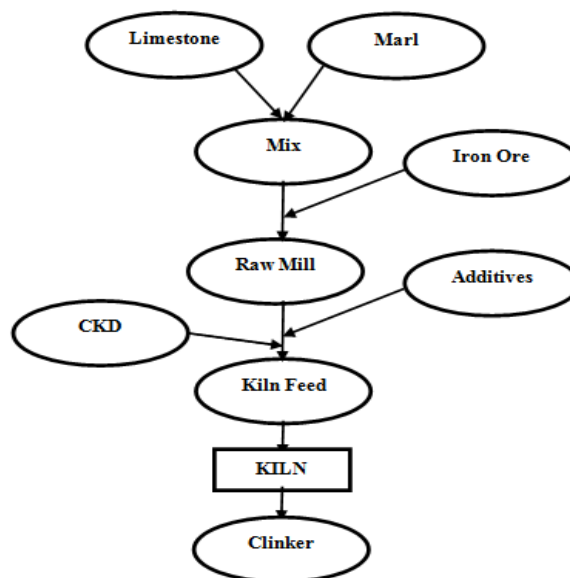


Fig.2 Simplified process schematic for mixing process of raw materials at Al-Mergheb cement plant [5]

Purpose of calculating the composition of the raw mix is to determine the quantitative proportions of the raw components, in order to give the clinker the desired chemical and mineralogical composition. For this, there are many methods of calculations. The basis of calculation is the chemical composition of the raw materials. So, the built program done based on (Alligation alternate method), this method allows the determination of the proportion of two raw material components. In this case, only the required lime content is fixed as a set point, so that the proportion of both components can be determined.

The main objective of mix calculation is preparation a kiln feed mixture with a suitable content of calcium carbonate (76-76.5%) to avoid hard burning process at high content of calcium carbonate content, as well as to avoid friction resulting from the silica on the formed coat on the internal kiln wall when calcium carbonate content is low.

B. Calculations and Simulation process

Depending on the schematic shown in Fig.2, a program built to calculation amount and determine type of additives (clay or limestone) will be add during preparation of kiln feed mixture. Algorithm of program is shown in Fig.3.

Basis of calculations depending on estimate of calcium carbonate content for raw materials, also proportion of mix depending on some of industrial data as following in Table 4.

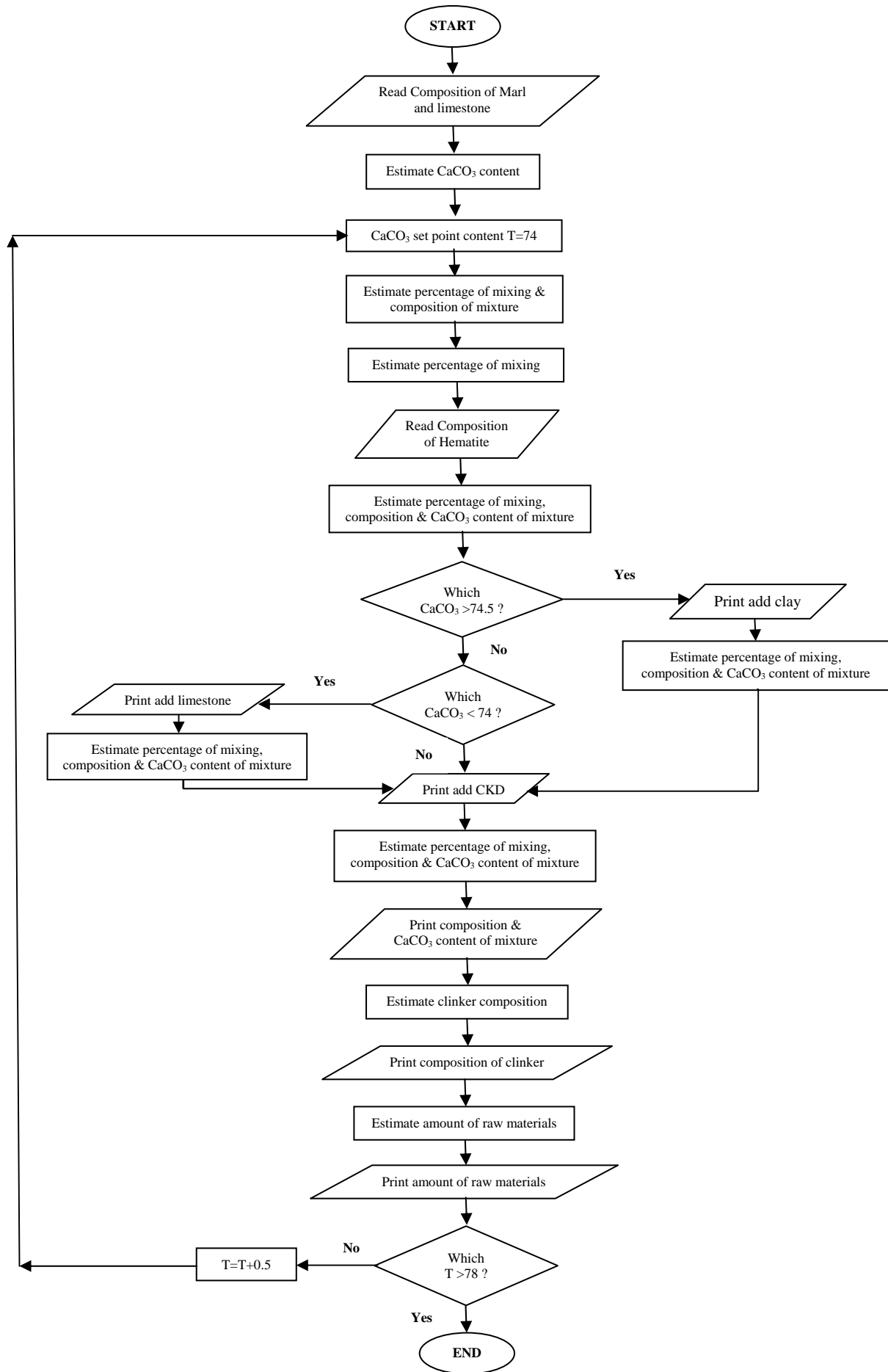


Fig.3 Algorithm of program

Table 4 Operating industrial data of plant [5]

Type	Amount	location
Amount of iron ore	2% wt. max. of inlet mixture Marl+Limestone)	Crusher feed
Amount of CKD	0.1% wt. max. of feed silos of kiln (kiln feed)	Feed Silos of kiln
Optimum kiln feed	76-76.5% wt. CaCO ₃ content	Kiln feed
Hard burning	> 78% wt. CaCO ₃ content	Kiln feed

C. Parameters of program

The main parameters effect on results which are the composition of raw materials, depending on this data set point of calculations for some parts of program changes to give the goal results (CaCO₃ content of kiln feed = 76-76.5) % wt. Quantity amount of raw materials fed to the process depending on amount of limestone that which is defined from the outset before you run the program within parameters as needed.

VI. RESULTS AND DISCUSSION

Run of program based on known of raw material compositions gave acceptable results, as shown in Table.5 results of some runs obtained from program running. The program give 9 runs based on the specified range (74-78)% with step size equal to 0.5.

Table 5 Some results of program

Data	Run 1	Run 4	Run 9
Set Point of Calcium Carbonate (%)	74	75.5	78
Amount of Limestone (Ton/hr)	55.3	55.3	55.3
Amount of Marl (Ton/hr)	30.19	24.28	16.06
Calcium carbonate content of mix (%)	74	75.5	78
Amount of Hematite (Ton/hr)	1.70	1.59	1.42
Calcium carbonate content of raw mill (%)	72.75	74.22	76.67
Material added	Add limestone	--	Add clay
Amount of Material added (Ton/hr)	15.39	--	2.56
Calcium carbonate content of raw mill after adding Material (%)	74.6	--	74.6
Add CKD	Add CKD	Add CKD	Add CKD
Amount of CKD added (Ton/hr)	1.025	8.8E-01	7.96E-01
Calcium carbonate content of kiln feed (%)	76.44	76.10	76.44
Total amount of kiln feed (Ton/hr)	103.61	88.91	80.45

Three cases are shown in Table 5, when limestone, clay and without additives are made. In each one calcium carbonate content of kiln feed not exceed basis range (76-76.5)% wt. Depending on data shown in Table 5, found that the first run are accepted industrial than other runs, due to provide a reserve of raw material by 50% more than the required entry into a kiln (feed rate to kiln ≈ 70 Ton/hr), giving the limit of safety during operation of the kiln to compensate losses or emergency states.

Composition of kiln feed calculated using program are shown in Table 6, for each run presents. In which note that CaO content of kiln feed (42.6-42.8)% wt. These values of CaO content within kiln feed produced are acceptable industrially due to lesser than 42.85% wt. that produce hard burning through kiln. As shown in Table 6 these values fit well with field data in last column.

Table 6 Chemical composition of calculated kiln feed and field data

Comp.	Run 1	Run 4	Run 9	Field data
SiO ₂	13.71168	13.79953	13.18194	14.43
Al ₂ O ₃	4.964175	5.01114	4.733855	3.9
Fe ₂ O ₃	2.669288	2.86015	2.777055	2.27
CaO	42.81046	42.62122	42.81046	42.61
MgO	1.23672	1.23976	1.189117	1.09
SO ₃	2.28E-01	2.34E-01	1.88E-01	0.1
K ₂ O	3.98E-01	3.96E-01	4.61E-01	0.9
Na ₂ O	1.24E-01	1.26E-01	1.21E-01	0.09
LOI	33.46112	33.30976	33.63758	34.61
Rest	6.25E-01	6.37E-01	1.088571	--
Total	100.2282	100.2337	100.188	100

Composition of clinker formed based on calculations are shown in Table 7, for each run presents. In which note that CaO content of formed clinker for these runs ranged (63.9-64.57)%wt. These values of CaO content within clinker produced are acceptable industrially due to lesser than 66% wt. in which a result of hard burning throughout the rotary kiln. As shown it close to field value 64.1% wt.

Table 7 Chemical composition of expected formed clinker and field data

Comp.	Run 1	Run 4	Run 9	Field data
SiO ₂	20.60702	20.69197	19.86356	22.22282
Al ₂ O ₃	7.460563	7.514050	7.133337	5.878449
Fe ₂ O ₃	4.011621	4.288718	4.18468	3.450678
CaO	64.33902	63.90923	64.51009	65.12717
MgO	1.858643	1.858991	1.791853	1.656632
SO ₃	3.429715E-1	3.504732E-1	2.833495E-1	0.152684
K ₂ O	5.975375E-1	5.939562E-1	6.941445E-1	1.374154
Na ₂ O	1.862408E-1	1.883E-1	1.819879E-1	0.137415
Rest	9.393549E-1	9.54786E-1	1.640342	--
Total	100	100.3505	100.2833	100

Many factors (modulus) refer to the quality of kiln feed and stability of clinker formed computed as shown in Table 8.

Table 8 Cement modulus of kiln feed, expected clinker formed and field data

Modulus	Run 1	Run 4	Run 9	Field Data	
HM	Kiln Feed	2.00563	1.966756	2.068853	2.068447
	Clinker	2.00563	1.966756	2.068853	2.064125
SR	Kiln Feed	1.796259	1.753147	1.755039	2.338736
	Clinker	1.79626	1.753146	1.755039	2.38209
AR	Kiln Feed	1.859738	1.752055	1.704631	1.718062
	Clinker	1.859738	1.75205	1.704631	1.703563
LSF	Kiln Feed	93.71548	92.46328	97.20356	92.00157
	Clinker	93.71549	92.46331	97.20356	91.52597

The hydraulic modulus of good quality cements were approximately 2 as shown in first raw and located in the permitted range (1.7-2.4). Silica ratio runs between (1.75-2.4), these values accepted in which that improves the burnability of the clinker and the formation of coating in the kiln. Data of alumina ratio runs in the acceptable range (1.5-2.5). Also, lime saturation factor of chosen run located in acceptable range (80-95).

All modulus estimated for kiln feed and formed clinker run acceptable and their accuracy of them closed to field data.

VII. CONCLUSION

A simulation of the mixing raw materials process and estimate composition of kiln feed, as well as formed clinker was made using the computer (Fortran 90 language) were done successful through fitting for the results obtained with field data of *Al-Mergheb cement plant*. Using this program can be avoid stopped of the plant when something defect goes in balance of raw materials or in the gamma-ray unit used to monitor the quality of feed to rotary kiln, through the permittivity range of results, which makes it easy to choose the specifications and quantities of raw materials required to preparing kiln feed at operating parameters selected. Also made it possible to prevent occurs of hard burning inside kiln and side adverse effects on kiln and other complementary units attached to it. It also enables to obtain quality of clinker within required specifications.

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