1. Introduction.

In the present time, clay bricks industry is considered one of the important strategic industries in Egypt referring to the need for huge amounts of the product for construction of new cities and establishment of new projects [1].

Shale / Clay deposits in various regions of the country represent the main raw material for clay bricks industry. They vary greatly in their mineralogical, chemical and particle size composition. Such variation affects considerably their behavior during forming and firing [2].

Clay bricks are made by shaping suitable clays and shales to units of standard sizes, which are dried and fired to temperature in the range of 750 to 950 ^{o}C [3].

Raw clay is composed mainly of clay minerals and non clay minerals. Clay minerals are composed mainly of hydrated aluminum silicate that are fine crystalline particles and develop plasticity when mixed with water[4]. The nature and properties of the clay minerals are determined to large extent by their crystalline layer structure which is based on the combination of silica tetrahedral layer Si_2O_5 joined with alumina octahedral layer $\{Al_2(OH)_4\}^{2+}$. Such a structure is generally classified into two main groups namely two layer structure e.g. Kaolinite group and three-layer structure, e.g. Montmorillonite and Illite groups [4,5]. The chief constituents of non-clay materials are quartz, feldspars, mica, calcite, dolomite and gypsum which represent the unaltered remainders of the parent rock (granite or pegmatite). Iron is usually present in clay deposits either in the form of free oxides, hydroxides, sulfides or carbonate or as lattice constituent in the clay minerals structure. Also, clay deposits frequently contain considerable amounts of organic materials [4].

Clay deposits are generally composed of sand, silt and fine particles. Particles size distribution of clay deposits lies in three sizes ranges of 2.0-0.063, 0.063-0.002 and < 0.002 mm, respectively with occasional coarser gravel particles > 2.0 mm [5]. Fig (1) shows particle size triangle of different types of clay deposits as well as particle size scales of various deposits components gravel, sand, silt and clay.

Clay performs two important functions in ceramic bodies.

a- Plasticity as a main Characteristic of the clay enables clay-water compositions to be formed and maintain their shape and strength during drying and firing. It is a basic property required for many of the ceramic forming processes commonly used.

  Plasticity is defined as the amount of water needed to be added to the clay batch materials for shaping. It can be determined by using pfefferkorn equipment as will be shown afterwards. Factors determining the degree of plasticity of a raw clay are mainly chemical and mineralogical composition and particle size distribution of the raw clay. With decrease of the particle size of the clay mineral, the required water amount for shaping the clay and consequently its plasticity increases.

b- Clays fuse over a temperature range depending on their composition in such away as to become dense and strong without loosing their shape [4-7].

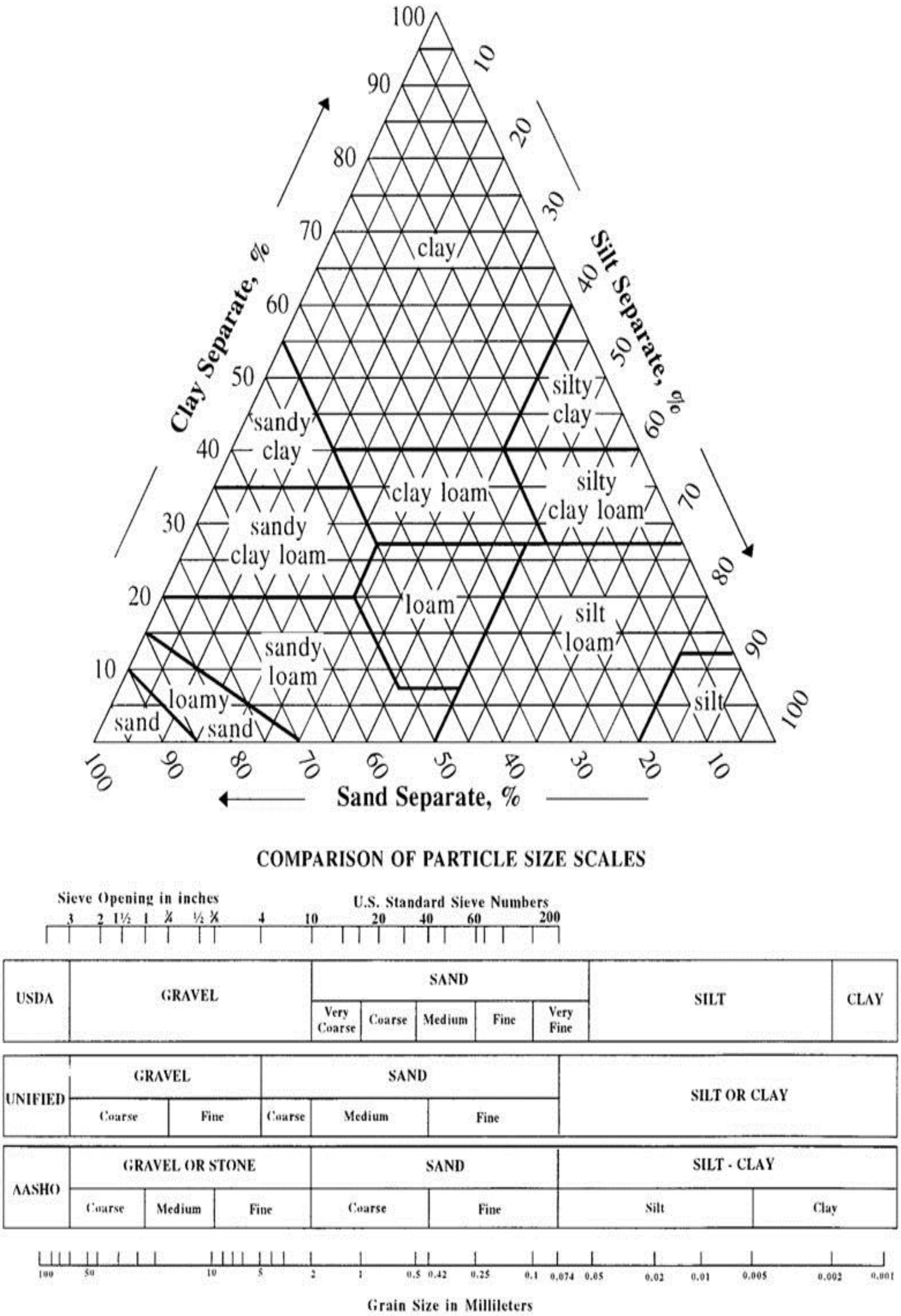

Fig (1): Particle size triangles of sand, clay and silt.

1.1. Clay minerals.

The combination of one silica tetrahedral layer Si_2O_5 and alumina octahedral layer $\{Al_2(OH)_4\}^{2+}$ as shown in Fig (2) leads to the structure of the kaolinite clay minerals $\{ Al_2(OH)_4 (Si_2O_5)\}$. The basal spacing of such a structure is 7.0 A^0. However, in the poorly crystallized kaolinite, some slight lattice substitutions of titanium and /or iron ions may occur which enlarge the basal spacing of kaolinite up to 7.20 A^0. In the structure of montmorillonite clay minerals, there are two silica tetrahedral layers existing above and below a central alumina octahedral layer as shown in Fig (2). The outstanding feature of the montmorillonite structure is that water and another polar molecule, e.g. ethylene glycol can enter between the unit layers causing the lattice to expand in the c- axis. The basal spacing of this structure is therefore not fixed and is ranging between 12.0 and 15.0 A^0 depending on the type of interlayer substitution. The structure of illite is similar to that of montmorillonite except that some of the Si^{4+} ions of tetrahedral layer is replaced by Al^{3+} ions and the resultant charge deficiency is balanced by fixation of K^+ ions in the interlayer spaces. Hence, the basal spacing of illite unit cell is fixed at 10.0 A^0 and the amount of its interlayer water is smaller than that of montmorillonite (7,8).

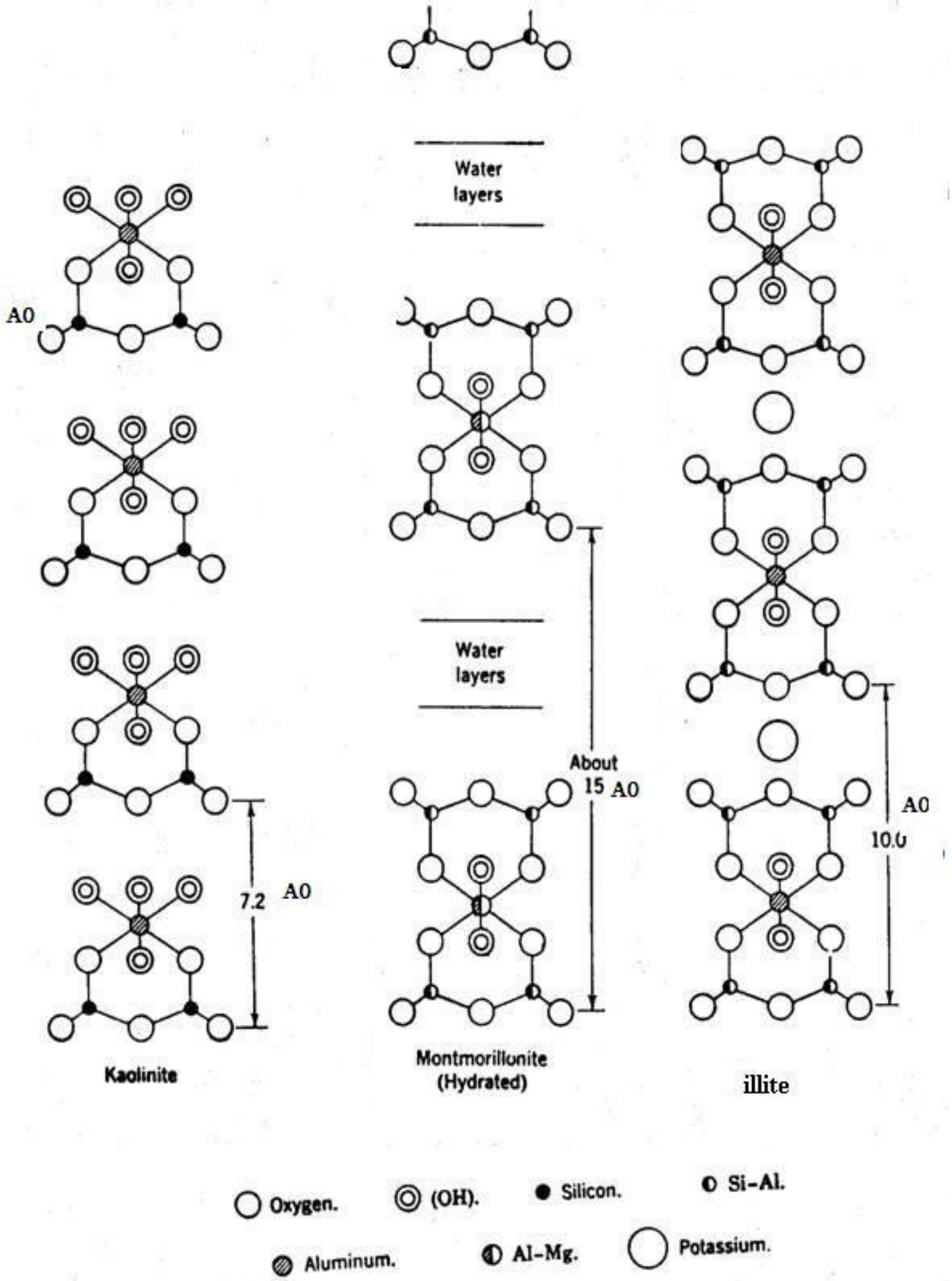

Fig (2): Layer structure of clay minerals

In clays the tetrahedral sheets are always bonded to octahedral sheets formed from small cations, such as aluminium or magnesium, coordinated by six oxygen atoms. The unshared vertex from the tetrahedral sheet also form part of one side of the octahedral sheet but an additional oxygen atom is located above the gap in the tetrahedral sheet at the center of the six tetrahedra. This oxygen atom is bonded to a hydrogen atom forming an OH group in the clay structure. Clays can be categorized depending on the way that tetrahedral and octahedral sheets are packaged into layers. If there is only one tetrahedral and one octahedral group in each layer the clay is known as a 1:1(tetra: octa) clay. The alternative, known as a 2:1 (tetra: octa) clay, has two tetrahedral sheets with the unshared vertex of each sheet pointing towards each other and forming each side of the octahedral sheet. Depending on the composition of the tetrahedral and octahedral sheets, the layer will have no charge, or will have a net negative charge. If the layers are charged this charge is balanced by interlayer cations such as Na^+ or K^+. In each case the interlayer can also contain water. The crystal structure is formed from a stack of layers interspaced with the interlayers [7-9].

Brief note about characteristic of some clay minerals are given in the following part.

1.1.1. Kaolinite.

Kaolinite is one of the most common clay minerals with the chemical formula {Al_2 $(OH)_4$ (Si_2O_5)}). It is a layered silicate mineral, with one tetrahedral sheet linked through oxygen atoms to one octahedral sheet of alumina octahedral. Rocks that are rich in kaolinite are known as china clay or kaolin.

Kaolinite has a low shrink-swell capacity and a low cation exchange capacity (1-15meq/100g). It is a soft, earthy, usually white mineral (dioctahedral phyllosilicate clay), produced by the chemical weathering of aluminium silicate minerals like feldspar. In many parts of the world, it is colored pink-orange-red by iron oxide, giving it a distinct rust hue. Lighter concentrations yield white, yellow or light orange colors [10-13].

1.1.2. Montmorillonite.

Montmorillonite is a very soft phyllosilicate mineral that typically forms in microscopic crystals, forming a clay. Montmorillonite, a member of the smectite family.

Chemically it is hydrated sodium calcium aluminium magnesium silicate hydroxide.

$$(Na, Ca)\ (Al, Mg)_6\ (Si_4O_{10})_3(OH)_6 \cdot nH_2O.$$

It is 2:1 clay, which means it has 2 tetrahedral sheets sandwiching a central octahedral sheet. The particles are plate-shaped with an average diameter of approximately 1 micrometre. Montmorillonite's water content is variable and it increases greatly in volume when it absorbs water. Its wet volume can be about 16 times larger than its dry volume.

Montmorilloniteite is a swelling clay, it is the main constituent of the volcanic ash weathering product, bentonite.

Potassium, iron and other cations are common substitutes; The exact ratio of cations varies with source. It often occurs intermixed with chlorite, muscovite, illite, cookeite and kaolinite [10-13]

1.1.3. Illite.

It is a non-expanding, clay-sized, micaceous mineral. Illite is a phyllosilicate or layered silicate; Its chemical formula is given as

$$(K, H_2O)(Al, Mg, Fe)_2(Si, Al)_4O_{10}[(OH)_2,(H_2O)]$$

It is characterized by considerable ion substitution. It occurs as aggregates of small monoclinic grey to white crystals. Illite occurs as an alteration product of muscovite and feldspar in weathering and hydrothermal environments [10-13].

1.2. Clay bricks manufacturing process.

Clay bricks are manufactured through the following main industrial steps: [14]

- Quarrying of raw materials.
- Preparation of the clay batch.
- Forming of extruded bricks.
- Drying.
- Firing.

Flow Sheet of the industrial processes in a clay bricks factory * is shown in Fig (3).

After all these operations, the bricks should be hard, not soften in water, nor spall under frost. [15] The produced brick should also satisfy the required specifications concerning: mechanical strength, dimensions, bulk density, apparent porosity, efflorescence. [15]

1.2.1. Quarrying of raw materials.

Concerning the clay raw materials used for bricks manufacture, the brick makers in the event of their clay being too lean confine themselves to remedying the situation by adding pure clay and if the clay is too fat they add sand or a very lean material . In some cases of good fortune these mixtures occur naturally [15].

***:** Company for manufacture and selling of clay bricks " Misr Brick "

1.2.2. Preparation of the clay batch.

The clay batch material is prepared for forming by being crushed in a crushing roll, tempered and mixed with water in a wet pan mixer consisting of a combination rolling and scraping action or in a pug mill in which knives are rotated through the clay + water mix as it passes through the pug mill trough. A substantial amount of air is added by either method and it is usual to pass the mixture through a vacuum de-airing chamber in which the material is shredded with knife blades or extruded through small openings in a vacuum chamber so that air in the clay can easily escape. The material is then ready for forming by Extrusion [14].

1.2.3. Forming of the clay bricks.

The most widely used method for forming clay bricks is extrusion through a vacuum auger .The stiff mud is forced through a die of hard steel or alloy by a motor- driven auger. Hollow portions can be formed by pins inserted in the die. Frequently steam or oil lubrication is used in order to decrease the force required and eliminate imperfections due to die friction. The die design is adjusted to give the minimum necessary resistance to flow and compaction.

The particular die design and degree of taper in the die vary substantially for different raw materials and die shapes. The degree of taper is best determined experimentally as the amount of taper which gives the maximum rate of extrusion for a given power input [14].

1.2.4. Drying.

Depending on the raw material and the shaping process, the moisture content of the ware after extrusion varies between 20-35 % [(15)] it must be dried before being fired at high temperature to form the final product.

Many of the defects which show up in the fabrication process such as the development of cracks and warping occur during drying. Some of these are caused by stresses induced during forming that only show up subsequently. However drying of aware of relative high moisture content, as in clay bricks industry, requires special care and control to avoid cracks due to excessive shrinkage [(16)].

In drying the clay ware [(14)], the initial drying rate is independent on the water content and depends solely on the temperature, humidity and rate of movement of the air over the surface of the ware. The drying rate is almost constant during this period, which constitutes the first phase of drying. At a water content such that the particles just come in contact, the water film disappears from the surface and the rate of drying decreases. That constitutes the begin of the second phase of drying where the drying rate decreases with time.

The major part of shrinkage occurs during the first constant rate period. The drying defects usually occur during the first period of drying. That refers to the differential shrinkage of the ware, where a solid structure free from water may be formed whereas the residual portion shrinkage is still going on [(15, 16)].

Generally the problems of drying increase with increase of the water content of the extruded brick and consequently with increase of its shrinkage extent. So non plastic materials, such as sand and grog materials, are added to the clay batch to decrease its plasticity and consequently decrease its water content and the extent of its shrinkage on drying (16).

So, it is necessary to control the drying rate in individual dryers especially in the shrinkage initial constant rate period. This can be achieved through:-

a- Controlling the humidity , temperature and flow rate of the drying medium (usually air)

b- Minimizing shrinkage of the brick during drying– mainly by minimizing the water content of the clay paste as possible. In other words controlling the plasticity of the paste, mainly by addition of opening agent such as sand, grog …etc (1).

Drying sensitivity of the extruded brick is assessed by plotting its shrinkage curve (Bigot Curve) (15). In such curve, the relative linear shrinkage of the extruded brick is outlined as a function of the percentage water removal. The gradient of the curve is a measure for the drying sensitivity of the raw material.

1.2.5. Firing.

After drying, the clay brick is fired to temperatures ranging from 700-950 ^{o}C, depending on the composition and the properties desired. During the firing process a viscous liquid is developed which allow the forces of surface tension to consolidate the ware and reduce porosity. The shrinkage which occurs in this process is equivalent to the porosity decrease. Control of the rate of firing is necessary since rapid shrinkage or contraction of surface can lead to sufficient stresses to cause failure. In addition, the time and temperature level of firing must be controlled to give a satisfactory and reproducible product. Clay bricks are normally fired in large tunnel kilns which are economical of fuel and have more or less satisfactory temperature control. The bricks loaded on a car pass through the kiln and the temperature distribution along the kiln is kept constant. Air for combustion is heated by cooling the fired ware through the cooling zone and the fuel combustion gases pass through the firing and preheating zones of the kiln and heat the ware entering. In this way the heated content of the combustion process is utilized with great efficiency (15, 16).

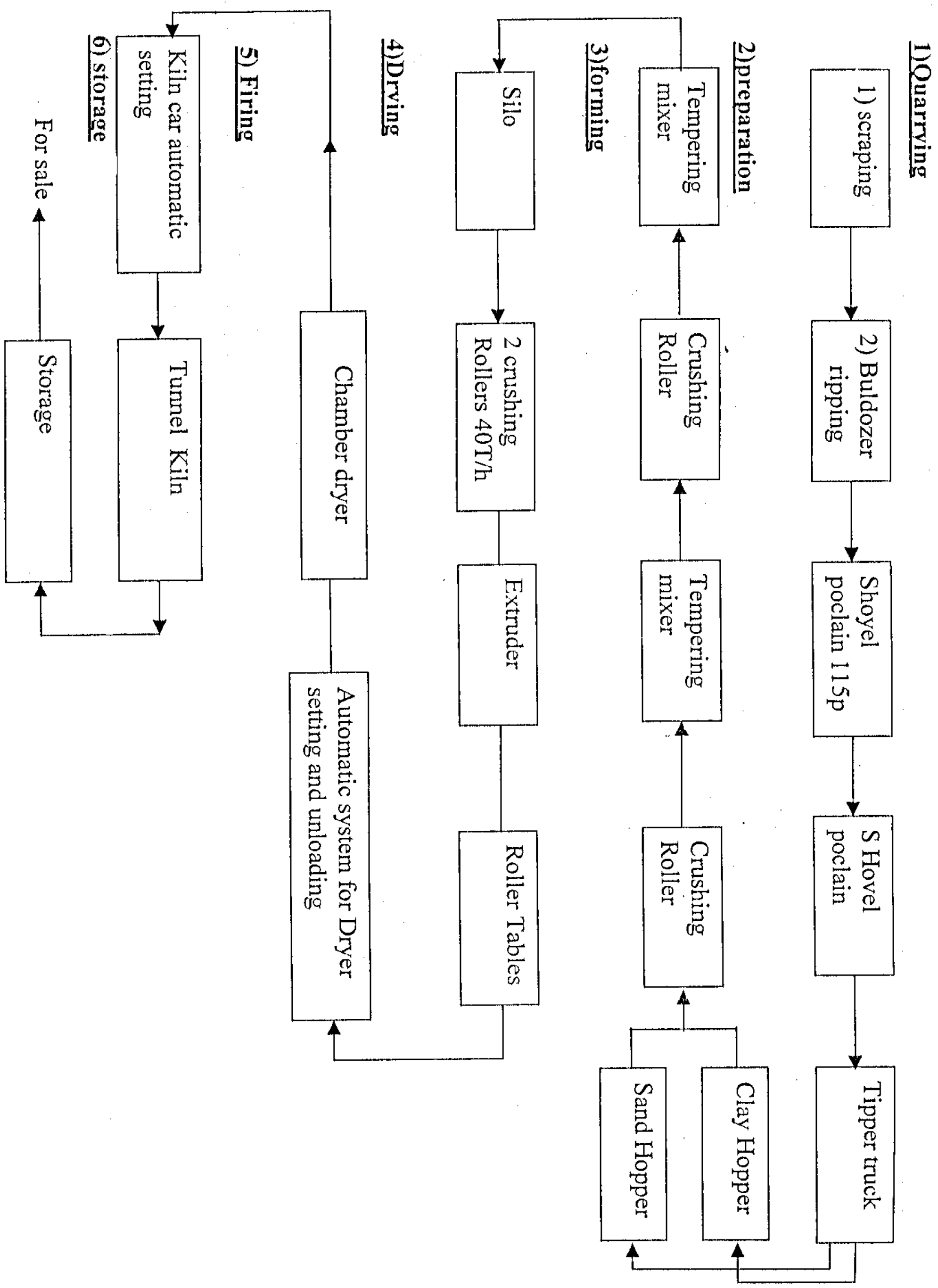

Fig (3): Diagrammatic representation of the manufacturing process.

1.3. Literature Review.

Long years ago, in Egypt, a number of authors have made a good characterization of clay and shale deposits distributed at many localities all over the country and have focused on the study of physico-chemical and ceramic properties of these deposits, in aim to evaluate its suitability and usability in manufacture of clay bricks [17-35] .

Gad and Abdul-Maksoud [17] studied the ceramic properties of kharaga clays for the production of mechanically formed building units. It was concluded that the addition of sand to the clay mixtures diminished the water workability and the drying as well the firing shrinkages to a considerable extent.

Janos [18] investigated the basic principle in testing the composition, ceramic and technological properties of clay bricks. It was concluded that the ceramic and technological properties of clay bricks are primarily determined by their mineralogical composition.

Galal [19] reported in details the compositional characteristics of Wadi El-Hai shale/clay deposits south of Helwan and their suitability for brick industry.

Ibrahim and Gad [20] studied the ceramic properties of clay deposits from both Mokattam area near Cairo and Tebbyne area near Helwan . It was concluded that both clays were suitable as substitute for the Nile-silt in manufacturing of clay bricks. Grog, sand and slag were added to overcome the drying and firing cracks of clay deposits from these regions. Briquettes were processed by the semi-dry-method under a pressure of 160-320 kg/cm^2 and fired at temperature ranging between 800-1150 ^oC. Samples fired at 800-900 ^oC and molded at 160 kg/cm^2 were quite good to serve as common red building bricks. Mixes molded under a pressure of 320 kg/cm^2 and fired at 1100 ^oC for two hours, are suitable for the production of engineering bricks, with 5.8 % water absorption and 438 kg/cm^2 crushing strength. Addition of 5% MnO_2 to the clay mixes fired at 1100 ^oC and soaked for two hours, results in lowering the water absorption of the brick to 2.2 % and increased its crushing strength to 637 kg/cm^2.

Ramez and Imam [(21)] reported and summarized the current Knowledge and situation regarding shale/clay deposits in Egypt in relation to the clay bricks industry.

Ramez and Sabrah [(22)] examined the ceramic properties of large shale / clay deposits found south of Helwan. They reported that the semi-industrial experiments made on studied clays proved its suitability for the building brick manufacturing.

Kamel [(23)] studied the pliocene sediments of Wadi El-Natroun, Egypt .He pointed to their possible utilization in building bricks industry. It was concluded that drying sensitivity as well as the drying and firing shrinkage are attributed to the structure of montmorillonite and its interaction with water.

Kebesh and Khalil [(24)] investigated three samples of Wadi Gharandel clay deposits for the production of building bricks, ceramic tiles, pipes, etc. It was found that the firing properties suggests that one of these samples could be used for the production of stoneware, mainly tiles , and the other two are suitable for building bricks .

Sabrah et al.,[25] investigated the physico-mechanical properties of Abu Zaabal basalt and Fayoum clay for the manufacture of building bricks. The mix composition with 75 wt % basalt and 25 wt % clay can be used for the manufacture of building bricks. It was concluded that the substitution of 10 wt % of clay or basalt by Tebbyne sand improves the bulk density, apparent porosity, water absorption and also crushing strength.

Sabrah et al .,[26] studied the ceramic properties of clay deposits from the Razonia area by Wadi El-Natroun, Egypt. The results obtained indicated the suitability of the clay deposits for the manufacture of clay bricks.

Helal [27] investigated the possibility of using clay deposits from Kom Oshim and Fayoum with many industrial waste products including grog, blast-furance slag and cement dust in the production of clay bricks.

Serry et al., [28] studied the effect of sand additions and firing temperature on the ceramic properties of clay of Wadi El Natroun . They prepared dense bricks by adding 40-50 wt % sand to the clay and also prepared vitrified ceramic tiles from the clays without flux addition by adding 50 wt % grog.

Ramez [29] compared the raw materials and mixtures used for making the old Egyptian red Bricks with that used in the clay brick industry . He reported a similarity in the composition of the used raw materials. It was recommended to use the Egyptian desert clay deposits in building clay brick industry.

Sharara [30] carried out the mineralogical and physico-chemical properties of high Kaolinite and montmorillonite – illite mixed layer Qatrani clay deposits, Egypt. It was concluded that these clays have enough plasticity. They can be easily formed using any shaping process. It was recommended that these properties are quite satisfactory for building clay bricks industry.

David A.Okongwu [31] studied composite clay brick containing various addition of saw-dust and spent lubricating oil (of SAE15-40 grades), then molded under a 15 MN/M^2 compaction pressure, dried, fired at 950^oC. Compressive strength, water absorption, and density measurement showed that small additions (<2 wt %) of sawdust led to improved Properties. Sawdust additions above this limit were deleterious, causing a progressive decrease in strength and an increase in water absorption capacity of brick. Oil addition, on the other hand, did not seem to have any useful effect on the characteristics of the brick.

El–Didamony, et al.,[32] reported the utilization of carbonaceous shale as a substitute for clay in clay bricks . The compressive strength and bulk density increases with clay content whereas absorption shows opposite trend. Also the liner shrinkage decreases of the carbonaceous shale content whereas. Also the clay could be substituted with 20-50 % carbonaceous shale to give clay bricks with reasonable properties.

Mandour and Mervat [33] studied two clay deposits located south of Helwan town area .They investigated their composition and properties in the view of mineralogy and chemical composition. For improving the ceramic properties of such clayey material, limited amount of sand and grog in different proportion were added to the clays to be suitable for clay bricks manufacture. Plasticity and firing behavior of the prepared mixes were also studied. The clay mixes were processed into building brick samples and fired up to firing temperature 1000 ^{o} C. It was concluded that wadi El –Hay clay cannot be used for building bricks manufacture without mixing it with Wadi Garawi and / or sand as well as grog. This is mainly due to its relatively higher content of clay fraction, which is composed of three layers clay minerals illite and smectite as well as its lower content of the non plastic minerals such as quartz and feldspar as compared with Wadi Garawi clay. This leads to excessive plasticity as well as drying shrinkage and strength. On firing Wadi El –Hay briquettes up to 850 ^{o} C, they show also higher total shrinkage and strength with minimum water absorption than Wadi Garawi. Hence, it was recommended to apply Wadi El-Hay clay as a limited additive up to 40 %, together with up to 20 % of each sand and grog to Wadi Garawi clay. This will lead to produce hollow bricks with reasonable shrinkage, strength and water absorption after firing the suggested batches up to 850 ^{o} C.

Farag, etal [1] carried out experimental as well as industrial filed studies for the optimization of local technologies for drying and firing of clay bricks.

Awad [34] studied the mineralogy and chemical composition of the shales related to Dakhla Formation near the Tushka area. The study revealed the presence of Illite, montmorillonit and kaolinite-montmorillonit mixed layer, in addition to kaolinite which is the major clay minerals. Moreover, the effect of sand and grog additives in different sizes and ratios on the shales of Tushka area was studied. It was concluded that, these additives reduce or prevent to great extent, cracking and bloating phenomenon of the fired briquettes. Adding sand decreases their crushing strength while adding grog increases the crushing strength of both solid and perforated briquettes made using Tushka shales.

Ismail Demir and Orhan [35] reported that the use of both coarse " passing through a 4.75 mm sieve " and fine " passing through a 600 µm sieve" crushed waste brick additives decreased the plasticity of the mixture of the clay. In order to contain comparable test results ratios of the waste (0, 10, 20 and 30% by mass) were added to the raw-clay. Also they reported that there are two factors affecting on the total shrinkage. These are the amount of the additives and the firing temperature. An increase of additives leads to a decrease of the total shrinkage and to increase of firing temperature leads to an increase total shrinkage. According to tests results, a mixture of up to 30 % fine waste-brick additives can be used in brick production. The most economical firing temperature was determined as 900 ^oC. Usage of waste material in the raw mixture minimizes the physical damage that occurs during brick production. The reuse of waste-brick material in brick production provides an economical contribution and also helps to protect the environment.

1.4. Aim of the work.

The present study aims at assessment of a raw clay from a quarry belonging to "Egypt Company for production and selling of clay bricks (Misr Brick)" as raw material for clay bricks manufacture. It aims also to raise the performance of the industrial process in the company by utilizing some wastes such as waste fired bricks (grog) and saw dust as batch additives with comparing the results with those of using sand as admixture. Misr brick is considered the largest company for clay bricks manufacture in Egypt.

It is noteworthy to mention that fired bricks wastes represent a huge environmental problem to the company. Their amount has been estimated as 350000U/year (1750 Ton / Year). Millions of tones are found over the ground floor of the company. That amount has been accumulated over the past twenty years. The saw dust was taken from the lot at wood-work refuse dump. Wood chips and strands were removed from the saw dust.