New technologies for production of building bricks and blocks without burning

Abstract The first three parts of this book address several new methods for the production of bricks and blocks, which are not subjected to high temperature firing. In the recent years the interest in the production of such building products has been growing because of the simplicity of the manufacturing operations and their good economic and environmental perfomance.

In the final, fourth part, our own study is presented, concentrated on the development of physical-chemical and technological basis of the method and composition for obtaining clay based building bricks and blocks without firing, including semi-pilot scale tests. The focus in the presented results is on the chemical problems in all processes as they are the base for obtaining product with good physical-chemical and physical-mechanical properties.

I. Guidelines for production of unfired building products (bricks, blocks, etc.) with high energy and environmental efficiency

One of the varieties of bricks, distributed earlier and today with limited application, is the one made only of clay. These building products have low strength and low durablity in humid environment, and therefore they can be used only for non structural walls.

Recently precast pressed clay blocks with improved design features and enhanced durability have been produced (Interlocking Compressed Soil Blocks - ICSB). To increase the strength and moisture resistance of the ICSB blocks, fly ash from thermal power plants (FA from TPP) or ground granulated blast furnace slag [GGBS] are added to the clay. However, traditionally, this type of bricks and blocks are stabilized with addition of 5 to 15% cement. These products have improved strength characteristics, moisture resistance and are of interest for the construction of housing in many countries worldwide such as China, India, Middle East, North Africa and others.

Many of the more advanced countries intensively develop a new scientific and technical direction by adding to the clay of lime and FA or GGBS. The binder phase in such products is generated by the chemical reactions of the clay material with additives and water, which typically take place at a temperature of 20 to 30ºC. The technology of this production stays simple and comprises the steps of mixing, moistening, plasticizing, mellowing, molding, curing in a humid atmosphere at room temperature and air-drying. Several such methods and technologies for the production of bricks and blocks are discussed. Some of these methods have been already implemented as industrial production, while others are being tested.

The main tasks that are intended to be solved by developing new methods and technologies are several:

- to obtain products with high performance comparable to those of the fired clay bricks;
- to lower expenses and save energy;

- to minimize the emission of harmful gases into the atmosphere;
- to utilize FA from thermal power plants and GGBS.

II. Unfired building products (bricks, blocks, etc.) obtained on the basis of FA from TPP

The book presents detail information about the quantities, composition and properties of FA, which are derived from coal burning in TPP. Data are also provided about the utilization of FA based on the experience in Bulgaria and worldwide.

The main problem, associated with the implementation of the FA as an artificial pozzolan, is its low rate of reaction with the lime. In this connection, the object of consideration are pozzolanic properties of the FA, which characterizes the ability of the active SiO_2 and Al_2O_3 of glass phase to react with the $Ca(OH)_2$ towards gel and crystal products, similar in composition and properties to those from the hydration of the hydraulic materials.

The review on the kinetics and mechanism of pozzolanic reaction FA-lime shows that the available data are too limited. This is a consequence of the complex qualitative and quantitative composition of the material {chemical, mineral (phase) and in grain size distribution}. The recently developed by Stoyanov and co-workers interesting method for studing the kinetics and mechanism of the pozzolanic reaction between Class F of fly ash and saturated aqueous solution of lime at temperatures of 20°C and 35°C is disscused. The rate constant of the reaction FA-lime at 20 °C is determined to be $Vr = 1.8x10^{-5}$ g $Ca(OH)_2/m^2.h$. At 35°C the reaction rate is limited by diffusion of lime reagent through the gel layer, deposited on the surface of the FA particles.

The analysis of the data, found in the literature on the kinetics of the reaction FA-lime, reveals several measures that could be taken to increase the rate of pozzolanic reaction in real conditions:

- Use of freshly precipitated $Ca(OH)_2$ as a reagent, because it lacks carbonation and the product possesses high reaction surface area;
- Long-term maintenance of high humidity in the systems composed of FA and lime for acceleration of the transport of the lime particles (molecules, ions) into the pores of the material;
- Use of FA with high content of glass phase, optimal grain size distribution and persistence of the chemical composition;
- For class C fly ash the content of reactive CaO is also important.

Technological difficulties in obtaining FA, which satisfy these requirements, are often significant. That is why the class F fly ash have found limited application in the production of unfired bricks and blocks, and relatively wider one as a mixture component for the production of fired at high temperatures building products.

The book covers two advanced technologies from the developed projects for production of unfired bricks and blocks, containing FA:

a. In Patent 2007 / 0000412 A1 an improved technology has been developed for the production of unfired bricks, blocks and other construction materials from class C fly ash with the introduction of air-entrainment agent, compaction at room temperature under pressure of 7 to 28 MPa and curing in humid environment. Chemical activation of the FA with lime is not applied because of their own binding properties. The newly developed product lasts 50 cycles of freezing / thawing, which satisfy the requirements for frost-resistance established with ASTM C 62 for firing bricks. The technology has been used for mass production in the United States.

b. Technology for production of unfired bricks based on class F fly ash, lime, gypsum, sand and calcium carbonate was developed in India (known as FAL - G Bricks). The problem in obtaining sufficient strength of the product is solved by combination of two processes taking place in the brick mass during curring: reaction of the FA with lime towards gel product and forming of ettringite. To increase the rate of the pozzolanic reaction FA with suitable properties has been selected and the molded products have been continuously saturated with water. The main process steps are shown in a flow chart of the FAL - G process. The strength characteristics of the obtained bricks or blocks are close to the lower limit of fired building products of this type, established by the standards of the respective country.

Results from other applied research, with no data for industrial implementations are also presented.

III. Unfired building products (bricks, blocks etc.) on the basis of clay, lime and GGBS

These building products are the result of detailed research on mixtures comprising GGBS as a component. The composition and the properties of GGBS are presented and a comparative analysis of pozzolanic properties of slag and FA is carried out:

The main phase in the GGBS, as in the FA, is a supercooled glass, determining the pozzolanic properties of the slag. The reaction between lime and GGBS gives silicate gel which contributes to developing a higher strength and durability of the products. Activation of the glass phase, however, occurs at lower pH values, below about 12, which gives an increased reactivity of slag in an alkaline medium and makes it more preferable than the FA for the purposes under consideration.

Factors influencing the reaction rate of slag-lime mixtures are chemical composition, content of glass phase and the reaction surface, the later being a function of the fineness of grinding (usually granulated material is ground to a particle size typical for cement). The slag can be activated in alkaline medium either by lime added separately or by the lime released during the hydration of cement.

Results from the study of two systems: clay – lime – GGBS-gypsum and clay – lime – GGBS are presented.

Studies of mixtures of clay–lime–GGBS–gypsum are conducted with kaolinite clay and did not lead to results having application as unfired building products. This is primarily due to the low rate of the reaction between kaolinite clay and lime and the consequent risk of delayed ettringite formation. Secondly, the final strength of the product is low (at age of 28 days it reaches 3 – 4 MPa).

The data about mixtures of kaolinite clay–lime–GGBS are more reliable. Based on this fact, many studies in the UK are concentrated on creating technology for unfired bricks. The optimal composition of the blend is 1.5 – 2 % lime, 5 – 5.5% GGBS, the rest up to 100 % being clay. The following technological operations are included in the scheme of the process: mixing, homogenizing, compaction of the wet material trough pressing (or extrusion), and curing of the building products in a humid environment at temperature of about 20ºC.

Under the defined optimal conditions bricks are manufactured in an industrial trial and after 90 days moist curing strength of 6 – 7 MPa is obtained. The other technical characteristics such as total water absorption, initial rate of water absorption and frost resistance are within the acceptable range compared to fired clay bricks.

An evaluation of the economic and environmental advantages of the technology is made, using the corresponding technical features of the production of fired clay bricks as a benchmark. Calculations have shown that the energy consumption for the production of unfired bricks from clay, lime and GGBS is 85 % lower, and harmful exhaust emissions are 80 % lower compared to the production of clay fired bricks. The same applies for the production of unfired blocks of clay, lime and GGBS.

IV. Unfired building products (bricks, blocks etc) on the basis of clay, lime and gypsum

This chapter covers:

Characteristics of fine grained soil materials suitable for lime and gypsum stabilization

The processes in clay-lime system are part of the processes in the more complex system clay-lime-gypsum. Therefore, the stabilization of mixtures of clay, lime and gypsum for production of unfired bricks, should also meet the criteria important for stabilization of clay with lime.

Subject of stabilization with lime are fine grained sandy and sandy-silty soils with moderate content of clayey substance and low in organic matter. In order to find the optimal composition and experimental conditions for carrying this process out, the following parameters have to be controlled: chemical and mineral (phase) composition; grain size distribution; basic engineering properties such as grading of the soil material, plastic properties, maximum density and optimal water content determined by Proctor test, and etc.

Kinetics and mechanism of the processes in the system clay-lime-gypsum

Knowing the kinetics and mechanism of the chemical interaction in the complex system clay-lime-gypsum-water, appropriate conditions for stabilization can be defined and properties of the products can be predicted. In this system both reactions clay minerals - lime and formation of ettringite are consecuitive, following the laws of the chemical kinetics, thus limiting the overall reaction rate by the rate of the slower one.

Clay minerals contained in the soil have a different reactivity towards lime. Their reactivity increases clearly in the order kaolinite → illite → montmorillonite. Among them, the kinetics of the kaolinite - lime reaction is studied better, because kaolinite forms well-shaped crystals and can be relatively easy obtained in pure form. The study of the kinetics of metakaolin-lime reaction is of great interest also, because metakaolin has a higher chemical activity than kaolinite, thus being closer to illite and montmorillonite.

Some results related to these properties of clay minerals are presented. They are obtained from some resent studies of the kinetics and mechanism of kaolinite-lime and metakaolin-lime reactions, carried out in a suspension of the material with a saturated $Ca(OH)_2$ aqueous solution, in the model test conditions. The particle size distribution of both materials is taken into account in the kinetic analysis, applying the method of Johnson and Mehl for presenting experimental data along with theoretical curves in dimensionless coordinates. The mechanism of the two reactions kaolinite-lime and metakaolin-lime is determined in a wide temperature range and the rate constant of the kaolinite-lime chemical reaction is found to be k = 2.10^{-11} m/s at 100°C.

The kinetics of formation of ettringite and mechanisms by which it causes swelling of building products are examined, using the detailed information about its properties in the literature. This analysis clearly shows that the reaction clay minerals-lime is the slower one and it limits the rate of the overal reaction in the system clay-lime-gypsum-water. There are to ways to increase the rate and reduce or prevent in this way ettringite expansion: stabilization of clays with high reactivity and addition of activators of the ettringite reaction.

The influence of various other factors for early completion of ettringite reaction is also evaluated. The more important are determined to be: accelerating of the dissolution of gypsum; the addition of 20 to 30 % more water than necessary (i.e., the optimal moisture content and the water needed for the chemical reaction in the system to take place); mellowing of the mass prior to molding of the products.

Experimental studies of different type of clays, directed to development of technology *for unfired bricks*

Experimental studies with several types of clays have been carried out: illite clay from the "Mirkovo" deposit near Sofia; loess clay from the town of Ruse region; and marl clay from Southwest Bulgaria. The role of different type of activators has been studied as well.

Stabilization of illite clay with lime, gypsum and activator. Bench-scale and semi-pilot trials

The clay from the "Mirkovo" deposit is sandy-silty, well graded. From the chemically inactive minerals it contain mainly quartz and plagioclase, and from the clay minerals - illite (hydromuskovite) 8 – 10 %, and vermiculite (a mineral from the group of montmorillonite) 5 – 6 %. The plasticity is moderate, PI = 15. The reactivity of illite clay relative to lime has been determined earlier experimentally using the dependence of the compressive strength on curing time of test samples. These data characterize the clay as a highly reactive. The kinetics of stabilization of the mixtures composed from illite clay – lime – gypsym – activator is studied using DTA/TG analysis, SEM and linear expansion measurements.

The following results have been obtained from the study of the composition and the properties of the clay, and the kinetics of the reactions occurring in the system clay – lime – gypsum – activator – water.

- The composition of the mixtures of illite clay, lime and gypsum is optimized - 7% of CaO and 5% gypsum hemihydrate are added to 100 parts by weight of clay. The activator is added in suitable proportions depending of its type. And 29 – 32 % water is imported also to the total amount of dry mass.

- Combined effect of the activators and the prolonged time of mellowing of the material before molding have been found to be responsible for early completion of the ettringite reaction. During this time the material is weakly bound and the etttringite expansion can take place without causing stress or with minimum stress.

- The optimal conditions of sample preparation have been determined to include the following operations: homogenizing and plasticization of the mixtures; mellowing of the plasticized material from 1 to 12 h; preparing of samples by pressing or extrusion; curing for 3 to 4 weeks in high humidity environment at 20-30ºC to raise strength; artificial drying at 30-35°C or air drying. The dried samples show volume consistency in wet and dry condition and keep their strength properties.

The bench-scale experiments for stabilization of illite clay – lime – gypsum – activator mixtures are carried out using the determined optimal composition and conditions. The produced samples are cube-shaped with 50 cm^2 wall surface. Compressive strength, initial rate of water absorption and total water absorption tests are performed. The compressve strength of wet samples immediately after removal from the moist chamber and of samples dried to constant weight is determined.

The compressive strength of dried cubes at the age of 28 days varies from 8 to 10 MPa, while wet cubes have strength 70 to 75% of that, obtained for the dry ones. The initial rate of water absorption is determined to be in the range 0.8 – 1.1 kg/m^2.min and the total water absorption 18 – 24 vol. %.

The semi-pilot tests aim to solve the following two tasks:

a. Development and testing of the operations constituting the flow-chrt of the process.

The basic equipment for these tests includes consecutively connected rolling mill, wet mixer and press type "Morando". Additional equipment is also used – dispensing system for the starting materials, moist chamber for the product.

The tests are conducted with series of 8 kg of clay each and the determined optimal amounts of chemical additives and water. After mixing, the mass is allowed to mellow for 6 h at ambient temperature (20 – 25 °C). One part of the plasticised material is extruded through a square nozzle to form dense samples used later for physical-mechanical tests; and the other part is mould through nozzle with cavity to obtain samples with about 40 % hollows.

The results from physical-mechanical tests of the extruded samples show that the expansion due to the formation of ettringite is from 1 to 2 % and this process is completed within the first 1 – 2 days after extrusion. The compressive strength is from 7 to 8 MPa, values which are slightly lower than those obtained in the bench-scale experiments. This is attributed to the surface roughness and small variations in the shape of the samples, caused by changes in the resistance of the leakage flow during extrusion. Water absorption of extruded material (12 – 14 %), is less compared to pressed cube material.

b. Preparation of bricks with dimensions 25/12/6.5 cm by pressing of the mass consisting of clay and chemical additives. The experiments were carried out with three types of mixtures: with actuator *AC3*; with actuator *AC4*; and without activator. The main component in the mixtures is clay from the Mirkovo deposit, and the determined optimal amounts of chemical additives and water are added to it. Pressing is performed in prefabricated wooden mold. Bricks are stored in a moist chamber at a temperature of 28°C, after being released from the mold. The following Table presents the results obtained for the strength of the bricks cured for 28 days.

Table: Strength characteristics of the bricks prepared with the addition of activators AC3 and AC4, and without activator

Type of mixture	**Series No**	**Addition of activator**	**Flexural strength, MPa**	**Compressive strength, MPa**
Mixture with Activator AC3	1	*AC3,* 5%	1.76 2.21	11.88; 11.79 11.62; 12.04
	2	*AC3,* 5 %	2.27 2.35	12.40; 12.33 12.56; 12.98
Mixture with Activator AC4	1	*AC4,* 20 %		10.50; 10.70 10.62; 10.84
	2	*AC 4,* 15 %		8.82; 9.10 9.03; 9.24
Mixture without activator	1			6.35; 6.12 5.81; 5.95
	2			6.07; 5.92 6.24; 6.05

The data presented in the table show that the strengths of the bricks prepared with 5 % of activator AC3 and 20 % of activator AC4 are higher than those obtained by the bench-scale trials. This is probably due to the better homogenization of the mixtures and the reduction of the mechanical loss of water during pressing. The high strength of the resulting bricks with real size makes them competitive to fired bricks from clay.

The semi-pylot tests confirm the main results obtained in the bench scale experiments and give opportunities to move to the next step of process development – conducting industrial trials.

Bench scale experiments for stabilization of loess with lime, gypsum and activator

The experiments are carried out with loess from the region of the Ruse town. Mineral analysis shows that the main phase in the loess is quartz, and its amount is about 65-70 %. The other components are mica, calcite, dolomite and plagioclase, listed as their content in loess decreases from 5 – 7 to 2 – 3 mass %. From clay minerals illite, montmorillonite and chlorite are found. Humus content amounted to 1.5 %. The loess has a relatively low plasticity, PI = 11. According to the classification based on particle size distribution and clayey substance content the studied material is defined by composition and properties as transitional type between typical loess and clayey loess.

The bench scale experiments of mixtures composed of loess, lime, gypsum and activator are carried out according to the procedures descibed for illite clay. The composition of the mixtures differs from those of the illite clay, due to the smaller amount of lime and water additions. (The additions of those components depend on the composition and the properties of the clay used).

Linear expansion of cubic shaped samples during curing and compressive strength, and total water absorption after drying are determined. The linear expansion behavior is close to that of the illite clay, but the height of the plateau remains slightly lower. The loess shows high strength at the age of 28 days (11 – 12 MPa) and moderate water absorption (16 – 19 %).

Bench scale experiments for stabilization of marl clay with lime, gypsum and activator

The clay originates from the region of Belo pole, located near to the town of Blagoevgrad, Southwest Bulgaria. Studies show that the clay is marl, sandy-silty, with moderate plasticity, IP = 15.

The mineral composition of clay characterizes it as a complex polymineral system - the main phase being quartz, followed by calcite and albite. Calcite is about 10 %, bound in the form of marl. The amount of the clay material is higher than that of illite clay and loess, composed of the minerals kaolinite, montmorillonite, hydromuskovite (illite) and chlorite.

Bench scale experiments are carried out under the conditions defined as optimal for the other two types of clay. Mixtures are prepared with lower water content (~ 25 %), because the fractions of sand and coarse silt dominate in the clay decreasing the consistence and causing difficulties in forming. This is the reason for the increase in the mellowing time of the material to 10 h.

Linear expansion measurements define marl clay as a low reactive. The expansion continues throughout the whole period of dilatometer measurements (one week), producing complicated experimental curves. This indicates association of clay minerals in the marl, which slows or blocks the pozzolanic reaction of clay substance with lime. The small amount of fine fraction in the starting material proves such mechanism of the process. The reason for the observed course of the experimental curves can be more complex, because such a continuous linear increase of the specimen dimentions is also characteristic for the clays with high kaolinite content.

Results for 28 days moist cured and dried afterwords cubes show low strength, as the average is between 3.5 and 4 MPa. The studies carried out with marl clay show it as inappropriate for stabilization with lime and gypsum for unfired bricks and blocks production.