Construction and Building Materials 61 (2014) 114–119

Contents lists available at ScienceDirect

Construction and Building Materials

journal homepage: www.elsevier.com/locate/conbuildmat

Ageing of clay and clay–tannin geomaterials for building

I. Keita [a], B. Sorgho [b], C. Dembele [a], M. Plea [a], L. Zerbo [b], B. Guel [b], R. Ouedraogo [b], M. Gomina [c], P. Blanchart [d,*]

[a] Laboratory of Physic and Chemistry of Materials, Bamako University, Mali
[b] Laboratory of Physic, Chemistry and Technology of Materials, Ouagadougou University, Burkina Faso
[c] CRISMAT, ENSICAEN, Caen University, France
[d] GEMH, ENSCI, Limoges University, France

HIGHLIGHTS

- Clay geomaterials for building are microcomposites materials with large sand grains embedded in a clay matrix phase.
- The macroscopic strength is from the binding property of the clays matrix and it can be increased with natural tannins.
- Ageing is evidenced by creep behavior under 0.2 MPa during 21 days, and is changed by water and tannin content.
- Ageing is induced by failing of microstructural internal bindings, leading to the weakening of the macroscopic strength.

ARTICLE INFO

Article history:
Received 12 November 2013
Received in revised form 3 March 2014
Accepted 5 March 2014
Available online 26 March 2014

Keywords:
Clay
Geomaterials
Tannin
Iron hydroxides
Parkia biglobosa pods

ABSTRACT

Clay geomaterials for building are clay sand mixture with controlled microstructural characteristics. The macroscopic strength mostly results from the binding property of clays and can be increased with the addition of various additives as tannins from plants. Such an improvement is due to the formation of chemical complexes tannin–clay–iron hydroxides. In this study, controlled clay–sand mixtures were used to obtain compressed blocks with a composite microstructure. The used clay is mixed with tannin compound that is simply obtained from *Parkia biglobosa* trees of Burkina Faso.

Creep curves during 21 days under a stress level commonly used in building (0.2 MPa) evidence a complex behavior with successive strain stages, depending on water and tannin contents. In general, creep is due to a slow and continuous volumetric deformation of the clay matrix and also to delayed micro-cracks propagation at sharp corners of sand grains. Ageing is evidenced by multi stage creep curves, resulting from local stress distribution within the matrix clay phase, which evolve with clay humidity and tannin content. Under a permanent stress, the microstructural internal bindings evolve with time leading to the weakening of the macroscopic material strength, which progressively tends to failure.

1. Introduction

Clay geomaterials for building are extensively used in either traditional or modern construction, but mechanical properties and ageing are key properties in use. They are mostly governed by the mineralogical composition of clays, by the content and size distribution of sands, and the role of moisture is also important. Consequently, properties for practical use of geomaterials for building must be investigated aiming at optimizing strength and durability, when building techniques and atmospheric conditions change significantly.

To increase strength in dry or wet conditions, clay–sand mixtures are often stabilized using cement additions (3–8 wt%). Usually, clay and cement are fine powders which cannot be easily and intimately mixed, leading to heterogeneous materials at the microscopic scale. It is a limitation of long time performance of blocks at the macroscopic scale, when compared with concrete blocks with similar cement contents [1]. Other synthetic products (alkaline silicates, metakaolinite...) have been extensively studied to improve bricks characteristics, but discussions remain about the effectiveness and the energy cost of these techniques [2,3].

Alternatively, stabilizers are obtained from naturally occurring substances and are used in traditional building techniques. In this study, we propose to use tannin compounds from a decoction of pods of Néré trees (*Parkia biglobosa*), which has been used in traditional construction and pottery techniques in Burkina Faso and

---

* Corresponding author. Tel.: +33 587502300.
E-mail address: philippe.blanchart@unilim.fr (P. Blanchart).

http://dx.doi.org/10.1016/j.conbuildmat.2014.03.005

Mali [4]. When mixed with mineral phases, tannins are dispersing agent for fine particles and change the rheology and plasticity of mixtures [5]. Tannins are also able to form chemical bonds with active sites on mineral surfaces leading to the change of the macroscopic behavior [6,7].

A typical microstructure of a clay geomaterials is in Fig. 1 (polished section after impregnation by the liquid polymer and polymerization). The SEM image (Secondary Electron Mode) shows the microcomposite aspect where large sand grains are embedded in the clay matrix phase, the content of the latter being the smallest. It is seen that grain size distribution of sands is optimized to increase the compactness of the materials after shaping by compaction, but remaining porosity coexist with the clay matrix.

Such compacted clay materials undergo creep against time under a constant pressure due to construction [8]. The extend of deformation depends on the matrix phase type, the grain size distribution and shape, and the porosity. Inelastic deformation often occurs by plastic yielding, which is controlled by the viscous characteristics of the clay matrix. In typical microstructure of Fig. 1, shear yielding is supposed to predominate [9], while volumetric yielding is not significant, due both to the small content of clay and the remaining porosity. But the behavior can be different with material containing rigid agglomerates which can be desegregated into small clusters under constraint. In that case, multiple local fracture processes in the clay matrix between large grains occurs with time, and the breakage of internal bonds leads to a macroscopic creep process [10].

The objective of this study is to investigate how compacted clay–sand bricks slowly deform under a constant load similar to that occurring in construction (0.2 MPa). Since these very simple materials are extensively used around the world, their mechanical properties must be improved to open new perspectives in building methods and in the durability of construction.

2. Materials and methods

The clay (named Koro) is mined in Burkina Faso (city of Bobo-Dioulasso). It is extensively used for pottery fired at low temperature (~800 °C) and also for molding bricks used for building after natural air drying. The clay was sieved at 63 μm to remove uncontrolled quantities of large grains of sands and gravels. Clay (18 wt%) was mixed with two sand grades of 40–160 μm (31 wt%) and 160–1000 μm (51 wt%). Relative quantities of clay and sands, and sand granularities are equivalent to average values used in Europe and Africa. Plastic pastes were obtained by mixing in a planetary mixer during 0.5 h, using 15 wt% of water, taking account the water content of the clay (2–4 wt%), which was preliminary measured. Plastic pastes were sieved at 1.6 mm to remove large agglomerates and die-pressed at 20 MPa to obtain cylinders of 50 mm diameter and 51 mm height. Water contents of samples were accurately controlled by frequent weightings during a slow drying at 45 °C and stopped when the targeted humidity was attained. A 1 wt% content of water was used to replicate dry building materials, and a 4 wt% content was necessary to replicate humid materials when a rainy atmosphere occurs. The volumetric mass of samples after pressing with 1 wt% of water is 1.98 ± 0.05 g cm^{-3}.

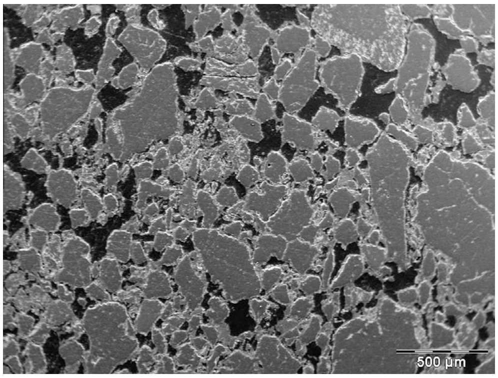

Fig. 1. SEM photo of a typical microstructure of a clay–geomaterial for building.

Tannin compounds were extracted from pods of Néré trees (*P. biglobosa*) [4]. They were preliminary dried at 50 °C, sliced and mixed with water (40 g of pods per liter of water) at room temperature during 3 days. After filtration, the dry matter concentration in the solution is 9.6 g/l. It was characterized by a Gallic Acid Equivalence method (colorimetric assay of phenolic and polyphenolic compounds) with the Folin–Ciocalteu reagent and UV spectrometry at 760 nm [11]. The concentration of tannins is 6.3 g/l, and this amount is relatively high in comparison to that obtained with different leaves of trees from African regions [12].

It is evidenced that *P. biglobosa* pod is a very interesting natural product since the high concentration of phenolic compounds containing hydrolysable tannins, which can be easily extracted by a simple and not costly decoction method at low temperature (30–40 °C). Complementary analyses of decoction by visible spectrometry (Lambda UV/Vis spectrometer and quartz cell) evidenced the predominance of hydrolysable tannins, mostly Gallic acid [8]. A more detailed study by HPLC spectrometry of the tannin composition is in progress, giving extended results that will be presented in a separate publication.

During experiments, the decoction was added at the planetary mixing stage, instead of water in clay–sand mixtures (15 wt% on the dry clay–sand composition). It gives a clay–Néré complex material after the same mixing and pressing stages than for clay materials, but continued by a curing time of 24 h at room temperature before the slow drying process, to attain the required residual humidity. In that cases, the added dry tannin compound is 1.44 wt%. The used clay–sand mixtures with the possible addition of tannin extract are in Table 1. Beside dried samples at 45 °C, a sample were subsequently heat treated at 100 °C during 4 h to favor the polymerization of the tannin extract from Néré [13]. During ageing and heating, both hydrolysable and condensed tannins polymerize into large, high-molecular weight complex tannins that strongly bind to iron species and precipitate as tannin–iron complexes.

The clay powders were characterized by X-ray diffraction on a Bruker AXS equipment (Cu kα; measurements in the 3–80 2θ° range, reflection mode on randomly oriented powder). Chemical analyses were performed by ICP (Iris Plasma Spectrometer). Microstructures of samples were observed by scanning electron microscopy (JSM-7001F Jeol) on polished sections after polymer impregnation, using secondary electron mode.

Stress–strain characterizations of shaped mixtures by pressing and drying were made by compression tests of cylindrical specimens, which were run at a crosshead speed of 1 mm/min at 22 °C, with an Instron apparatus equipped with a 10 kN load cell. The accuracy of all stress values is ±0.1 MPa. Creep tests of cylindrical specimens were run at 22 °C under 0.2 MPa constant load during 21 days, to characterize the long term strain of geomaterials. The strain was recorded periodically (5 min) with a 0.1 μm accuracy. The used 0.2 MPa constant load is equivalent to that of a 8 m high vertical wall (i.e. two building floors).

3. Results

The chemical composition of the Koro clay (Table 2) is similar to that of commonly encountered clay with high iron content. The powder XRD pattern is in Fig. 2 showing that the major minerals are the sheetsilicates smectite, illite and kaolinite, associated with quartz and a small quantity of orthoclase feldspar. Significant quantities of iron minerals are clearly identified as hematite Fe_2O_3 and goethite $Fe^{3+}O(OH)$. Using the chemical composition of Table 2 and the mineral phases identified by XRD, the quantitative mineralogical composition was calculated (Table 3). For that calculation, we considered that chemical compositions of each mineral phase are close to ideality, with no structural iron in silicate minerals. It shows that phyllosilicates minerals predominate (54 wt%) and that the relatively high quantity of both hematite and goethite (6 wt%) is a specific characteristic of this clay.

With the clay–Néré complex, the X-ray pattern is compared with that of the clay alone in Fig. 2. There are no significant differences in position and intensity of montmorillonite and kaolinite peaks, relatively to quartz that can be supposed to present a low

Table 1
Material compositions, humidity content and possible heat treatments.

Materials	Drying temperature (°C)	Names
KORO-1% humidity	45	K1
KORO-4% humidity	45	K4
KORO + tannin-1% humidity	45	Kn1
KORO + tannin-4% humidity	45	Kn4
KORO + tannin-100 °C	100	Kn1-p