Ovalbumin as natural organic binder for stabilizing unfired earth bricks: Understanding vernacular techniques to inspire modern constructions

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Highlights

Ovalbumin strongly increases the compressive strength of earth bricks.
Ovalbumin significantly improves the resistance of earth bricks to water.
Ovalbumin does not modify the thermal conductivity of earth bricks.
Ovalbumin decreases the MBV of earth bricks slightly.
Ovalbumin is more efficient than cement or hydrated lime for earth stabilization.

Abstract

Cement and hydrated lime are usually effective for stabilizing unfired earth but they lead to a significant CO_2 footprint. In the search for sustainable, efficient alternatives, old and vernacular techniques may provide ways of replacing such mineral binders. The example of the use of chicken egg white protein (ovalbumin) to improve earth construction materials is addressed in the present case study. It was used to stabilize two soils with different mineralogical compositions. Addition of 2 wt % and 4 wt % ovalbumin strongly increased both the dry and wet compressive strengths (water resistance) of the soils. For these properties, ovalbumin was a more efficient stabilizer than cement or hydrated lime. Concerning hygrothermal properties, the thermal conductivity of the soils did not change with stabilization, while the moisture buffer value decreased slightly but remained at least "good" according to the Nordtest criterion. Microstructural analysis can explain the efficiency of earth stabilization using ovalbumin by the formation of a gel that fills the microcracks in the soil and strengthens it by gluing them up.

Keywords

Earth bricks; stabilization; ovalbumin; compressive strength; water resistance; hygrothermal properties.

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1. Introduction

The literature of recent decades records a profusion of research on building with unfired earth. This trend is favoured by the context of climate change challenges that result in unfired earth being considered as a serious alternative to traditional cement concrete as a sustainable construction material because of its social, economic, environmental and hygrothermal qualities. Ancient and vernacular earth constructions around the world provide undeniable evidence (Bahobail et al. 2012) that a material that is available everywhere and can be used with little or no transformation will be affordable for populations in developing countries and cost-saving for people in developed ones. Except for possible mechanical extraction, transport and building operations, almost no CO_2 footprint is imputable to this material. Furthermore, its hygroscopic performance and the thermal inertia it induces can contribute to energy saving in modern buildings, where artificial ventilation is necessary to ensure users' comfort (Morton et al. 2005; Houben et al. 2006; Gernot Minke et al. 2009).

Thanks to the commitment of scientists and some stakeholders, a broad public is nowadays being encouraged to build with earth on the basis of the qualities mentioned above. Nevertheless, current construction standards, strongly inspired by half a century of massive use of cement concrete, remain too challenging for such natural material. The mechanical and durability performances of unfired earth are basically the parameters that limit its ability to compete with cement concrete. Regarding the compressive strength, Houben et al. (2006) have pointed out that, with its common dry compressive strength of around 2 MPa, earth is suitable for 2 storey buildings, where the compressive strengths of tens to hundreds of mega pascals available with cement concrete are not necessary. However, its hardening mechanism, which depends on the matric suction of water between clay sheets, is reversible. So, when exposed to liquid water, the material loses its mechanical strength, threatening the structure (load bearing walls) with collapse. To tackle this problem, construction intelligence, such as the use of waterproof substructures and roof extensions, has often been sufficient, as shown by the vernacular construction heritage in numerous countries (Houben et al. 2006; Gernot Minke et al. 2009; Aubert et al. 2019). However, in some extreme situations, like accidental water damage or flooding, which are likely to occur due to heavy rains resulting from recent climate change, earthen parts may be exposed to liquid water despite the protective techniques. In these cases, it could be necessary to ensure a minimum wet compressive strength to guarantee users' safety. Moreover, for industrial use, earth structural subunits may be transported from the plant to the construction site. Usually, this induces higher mechanical loads than those encountered in on-site use, so the minimum mechanical strength requirement may have to be increased in consequence. Finally, once in place, the corners of unfired earth walls are known to be sensitive to abrasion or fragile with respect to adverse shocks.

For all these reasons, most research has used large amounts of cement and other artificial binders, such as lime, to stabilize earth materials and improve their mechanical and durability performances. Unfortunately, the mechanical and durability performances obtained remain far lower than those of cement concrete materials and the carbon footprint induced in unfired earth materials by using mineral binders inhibits their sustainability (Van Damme et al. 2017). This issue has been addressed in a previous study on the effects of small amounts of cement and hydrated lime additions on the properties of two different soils (Ouedraogo et al. 2020). We recall that the paper's hypothesis was that the maximum mineral binder contents for earth brick stabilization with a low CO_2 footprint were determined as 4 wt %, by considering an equivalent cement content in a conventional Hollow Concrete Block (HCB), which contains 50 % of voids. In that study, 2 wt %-4 wt % of mineral binders (cement and lime) were used to stabilize two soils with different mineral compositions. The results obtained on both soils showed that the dry compressive strengths of the soils did not change unless the manufacturing density was increased. A significant improvement of the wet compressive strength (water resistance) was recorded, especially with 4 wt % addition of cement. The hygroscopic performances of the soils decreased but remained at least at a "good" level. When compared to HCB, the relevance of this stabilization of the soils with 2-4 wt % of mineral binders is still a subject of discussion. Although the CO_2 footprint was related to the use of the mineral binders, the dry compressive strengths of stabilized earth materials remained lower than those of HCBs, and their water resistance was far below the HCBs'. Only the hygroscopic performances were better than in HCBs.

Therefore, the search for alternative binders with low environmental impact turned towards old and vernacular stabilization techniques using natural organic binders from animal and plant by-products (Camões et al. 2012; Vissac et al. 2017; Bamogo et al. 2020). To date, scientific works on these types of binders remain sparse in comparison with the studies conducted on the use of mineral binders for earth stabilization (Danso et al. 2015). Various natural organic polymers, such as gluten and casein (Guerrieri et al. 2012), alginate (Galán-Marín et al. 2010; Pinel et al. 2017), chitosan (Aguilar et al. 2016), xanthan gum (Chang et al. 2015), carrageenan (Chang et al. 2015), tannin (Sorgho et al. 2014), starches (Alhaik et al. 2017) and cow dung (Millogo et al. 2016, Bamogo et al. 2020) have been used to stabilize earth for construction. It is noteworthy that most of them were used as water repellent coatings or plaster mixtures rather than being incorporated in the walls.

A sorting test presented in another study (Ouedraogo et al. 2019) identified the potential of ovalbumin (chicken egg white protein) as a promising binder for earth stabilization. The main protein of chicken egg white is albumin, a water-soluble protein found in the serum of animal blood or in egg white (ovalbumin) (Gooch 2007). According to Mine (1995), ovalbumin accounts for about 54 % of the mass of egg white proteins. It is a globular phosphoprotein in which half of the amino acids are hydrophobic. Ovalbumin also contains free sulfurdryl groups that lead to the formation of gel when heated (Mine 1995). Other proteins, such as ovotransferrin (12 %), ovomucoid (11 %) and ovomucin (3.5 %) are also found in egg white. Egg white proteins, including ovalbumin, are known to be highly surface active thanks to their amphiphilic property. They are therefore used in various industries for the formation of stable foams (Phillips 2009). Literature on the use of ovalbumin for earth stabilization is sparse but Vissac et al. (2017) reported its use in vernacular plaster recipes for protecting earth walls. The only available work on ovalbumin used as a binder in construction concerns animal blood albumin (Winkler 1956; Abdelhadi et al. 1998; Fang et al. 2015). The main consumption of egg white is recorded in the food industry. Nevertheless, other sectors use its physicochemical properties. For instance, it is used for the formation of ceramic foams (Liu et al. 2019).

The present work studies the effects of ovalbumin addition on the microstructural transformations and the physical and mechanical properties of earth bricks made with two different soils having different mineralogies. These soils were characterized in depth in a previous study (Ouedraogo et al. 2020). In the work presented here, the effects of ovalbumin additions on mechanical performance and resistance to water were evaluated using the dry and wet compressive strength of stabilized earth bricks. In parallel, the effects of this stabilization on the hygrothermal properties of earth bricks were studied using thermal conductivity and Moisture Buffer Value (MBV). Finally, in order to better understand the mechanisms at the origin of the stabilizing effect of ovalbumin on earth materials, the mineralogical modifications of stabilized bricks were studied using X-Ray Diffraction (XRD), infrared spectrometry (IR) and Scanning Electron Microscopy (SEM). This paper aims to appraise the potential of ovalbumin as an environmentally friendly binder for earthen construction. At this stage, no direct industrial application is considered because of the ethical issue arising from the fact that eggs are among the most valuable food products. Furthermore, the goal of the case study is to understand the stabilization mechanism of the ovalbumin, which may be applicable to more available bioproducts. In the discussion part, the performances obtained with ovalbumin will be compared with those obtained using cement and lime stabilization. In particular, the performances of ovalbumin-stabilized earth bricks will be compared to those of hollow concrete blocks, which can be considered as the reference for limiting binder content.

2. Research aim

The case study aims to assess the effectiveness of using natural biopolymers to stabilize earthen constructions, as various reviews have reported their use in vernacular construction techniques around the world for the purpose of protecting earthen buildings against weathering. In the global warming context, building modern constructions with earth is attracting increasing interest from industrials because of its low environmental impact and its social and economic advantages. However, earthen construction is challenged by current modern construction standards: high mechanical strength requirements, water resistance etc. For this reason, cement and lime are widely used for stabilizing the earth despite their CO_2 footprint. Albumin is a protein that is mainly available in animal blood and in egg white (ovalbumin). Its use as a water repellent on cultural buildings has been reported. This work highlights the efficiency of ovalbumin as a natural organic stabilizer in earthen construction materials by comparing it with cement and lime and seeks to understand the underlying mechanisms so that they can be extended to other organic biopolymers.

3. Materials and procedures

3.1 Raw materials

3.1.1 Ovalbumin

The product used for this study was supplied by the certified chemicals company Sigma Aldrich. Ovalbumin is obtained in powder form by lyophilizing chicken egg white. The ovalbumin amount determined by agarose electrophoresis test ranges from 62 to 88 wt %. The constitutive water makes up 10 wt % and the nitrogen content is 12.5 wt %. Ovalbumin has a water solubility of 50 g/l. All characteristics mentioned were taken from the supplier's technical sheet.

3.1.2 Raw soils

The two soils used for the study were provided by traditional brick factories in the south of France. They were chosen for their differences of mineralogical composition, in order to highlight its impact on the stabilization efficiency. Their characterization was performed in a previous study (Ouedraogo et al. 2020). Table 1 recalls the main geotechnical characteristics of the two soils.

Table 1: Geotechnical, chemical and mineralogical characteristics of the raw soils B and N (Ouedraogo et al. 2020)

Table 1 shows that both soils had similar particle size distributions and could be classified as fine soils. However, they showed significant differences in their mineralogical compositions. Besides the quartz, the feldspars and illite, each soil contained other specific minerals. Soil B was composed of kaolinite, muscovite, goethite and calcite, whereas Soil N consisted of montmorillonite, chlorite and calcite. This difference induced different geotechnical behaviour between the two soils, in particular when the methylene bleu value was considered. The methylene bleu activity index (A_{CB}), computed as the methylene blue value over the clay content was 8.3 for Soil B and 18.2 for Soil N. This parameter shows the water affinity (A_{CB}) of the predominant clay minerals in the soil's clay fraction. With A_{CB}=18.2, Soil N was considered to be very active, while Soil B was moderately active, with A_{CB}=8.3 according to the classification of Lautrin (1989), quoted by Chrétien et al. (2007). The differences of mineralogical compositions between the two soils were expected to influence the stabilization effectiveness and they were chosen for that purpose.

3.2 Procedures

3.2.1 Specimen manufacture

Two types of specimens were made by static compaction: cylindrical specimens 5 cm in diameter and 5 cm high (Φ5H5) for dry and wet compression tests and parallelepipedal specimens 15*15*5 cm^3 for hygrothermal tests.

Cylindrical specimens (AFNOR NF EN 13286-53 2005, 13286–53) were manufactured by static compression using a hydraulic press with a load capacity of 2.5 tons. The soil was pre-wetted to a moisture content of about 10 wt % and kept in a hermetically sealed bag for 24 hours. The aim of this step was to ensure better absorption and distribution of moisture in the soil. The actual moisture content of the pre-wetted soil was measured according to the standard (AFNOR NF P94-050 1995) to determine the amount of water to be added in order to reach the moisture content required at the time of manufacture. The additional water and the required amount of binder were then mixed with the pre-wetted soil before moulding. The ovalbumin and the mineral binders (cement and hydrated lime) were mixed at the rates of 0, 2 and 4 wt % with the Soils B and N for specimen moulding. Because the amount of ovalbumin was limited, no test was performed to assess its effect on the soils' optimum Proctor densities. Therefore, all the specimens were pressed at the same water content and the same dry density as the optimum Proctor values for raw soils (w=14.1 wt % and ρ_d=1.88g/cm^3 for soil N and w=15.6 wt % and ρ_d=1.86g/cm^3 for soil B). The specimens were then dried to constant weight (Δm_{24h}<0.1 wt %) at the room temperature of 20 °C and relative humidity of 50 % before the performance tests. The cement and lime stabilized specimens were tested at the age of 35 days (including 21 days of curing in hermetic bags and 14 days of drying) while the ovalbumin stabilized specimens and the unstabilized specimens were tested after 14 days in a drying room.

3.2.2 Dry compressive strength and elastic modulus

These tests were carried out on cylindrical specimens using a hydraulic press with a force transducer having a capacity of 50 kN at a loading speed of 0.2 kN/s, corresponding to 0.10 MPa/s for a section of 50 mm. A press plate displacement sensor enabled an average deformation of the specimen to be computed during the compression test. The displacement sensor recording was triggered by the onset of load sensing. Hence, the approximate elastic modulus of the specimens was determined as shown in Fig. 1a and Fig. 1b.

Fig. 1: Mechanical measurements
(a) : Compressive strength; (a and b): Elastic modulus

More accurate techniques are mentioned in the literature for measuring the deformations of the specimens, with linear variable derivative transform (LVDT) sensors (Lima et al. 2012; Bui et al. 2014; Miccoli et al. 2014) However, the method used here permitted the effect of stabilization on the materials' stiffness to be identified in a comparative manner.

3.2.3 Water resistance / wet compressive strength

The water resistance of the samples was evaluated by using the wet compressive strength test according to the French CEB standard (XP P 13-901 2001, 13–901). The specimens were immersed in a water tank for 2 hours, then removed and wiped with a damp towel. They were finally placed in an airtight bag and stored for 48 hours in the room at a controlled temperature of 20 °C. The compressive test was then carried out according to the procedure presented in the previous subsection.

3.2.4 Thermal conductivity

The thermal conductivity of the samples was measured using a λ-Meter EP 500 according to the guarded hot plate method. Another measurement by the hot-wire method was carried out using a hot-wire conductivity meter. The latter method is known to be rapid but less accurate than the former. Results of the two methods were then compared. Before the test, the specimens were oven dried at 50 °C until they reached constant weight (7 days) and cooled down in a super drier at a temperature of 20 °C (3 days).

3.2.5 Moisture Buffer Value (MBV)

The water buffer capacity was conducted on $15*15*5cm^3$ specimens according to the NORDTEST protocol (Rode et al. 2005). To meet the penetration depth at 1 % relative humidity ($dp_{1\%}$) criterion, specimens were prepared in a particular manner. Based on data from the literature (Abadie et al. 2009; Moevus et al. 2013; McGregor et al. 2014), the greatest penetration depth at 1 % of RH of the most hygroscopic raw earth was determined to be about 5 cm. So the specimens were sealed with aluminium tape, except on the two opposite parallel faces, which were 15 cm apart. Therefore, the mid cross-section was at 7.5 cm from the exchange areas, a distance greater than the maximum $dp_{1\%}$ of 5 cm. In addition, the sum of the two exchange areas exceeded the 100 cm² needed to guarantee sufficient water uptake to calculate the MBV.

3.2.6 Chemical and mineralogical analyses

For the following tests, an agate mortar and pestle was used to grind the samples into a powder that passed through an 80 µm sieve. XRD was performed on a BRUCKER D8 ADVANCE 5000 diffractometer equipped with a SOLLER rear monochromator and a copper anticathode (CuKα). Infrared diagrams were obtained using the Perkin Elmer UATR1 Frontier FT-IR spectrometer on powders of samples crushed to 80 µm in an agate mortar.

The microstructure observations were performed using a JEOL JSM 6700F scanning electron microscope (SEM) coupled with an energy dispersive X-ray spectroscopy (EDS) analyser. Pieces taken from broken specimens of the dry compressive test were polished (“polished specimens”) or left with rough surfaces (“fractures”). They were then carbon coated for SEM-EDS analysis.

4. Results and discussion

4.1 Effects on mechanical strength and durability

4.1.1 Dry compressive strength

The effect of ovalbumin addition on the dry compressive strength is presented for both soils in Fig. 2.

Fig. 2: Compressive strengths of the ovalbumin stabilized soils B and N

Fig. 2 shows that, when the soils were mixed with 2 wt % and 4 wt % of ovalbumin, the dry compressive strengths almost doubled for Soil N (+98 % to +135 %) and tripled for Soil B (+177 % to +281 %). These marked enhancements originated from various chemical properties of ovalbumin. First, its water solubility allows it to penetrate the soil matrix uniformly for a better interaction with its particles. In addition, thanks to their amphiphilic properties, the hydrophilic poles of ovalbumin molecules adsorb on the -OH sites available on the particles of the clay surface to glue them together strongly (Anger et al. 2013). It is likely that these adsorptions result from the hydrogen bonds between organic polymers and clay platelets (Ledoux et al. 1966; Lagaly et al. 2006; Chiu et al. 2014). The improvement varies according to the soil nature. In fact, the raw compressive strength values of Soil N are higher than those of Soil B while the improvement rate is greater for Soil B than Soil N. This could come from the difference in mineral contents between Soil N (illite, chlorite and montmorillonite) and Soil B (illite and kaolinite). Drying cracks occurred only on Soil N mixed with 2 wt % and 4 wt % ovalbumin as shown in Fig. 3.

Fig. 3: Occurrence of drying cracks on specimen of soil N stabilized with ovalbumin

This was probably due to montmorillonite related swelling-shrinkage combined with the well known surfactant effect of ovalbumin (Phillips 2009; Anger et al. 2013), which acts as a repulsive force when the mixture is in the wet state. The cracks thus created reduced the effectiveness of the binder on Soil N dry compressive strength.

To date, few works are known to have been conducted on earth stabilization using ovalbumin for building purposes. Most of them deal with the use of the mixtures as plasters or the use of ovalbumin as a surface water repellent (Vissac et al. 2012). Therefore, there is no reference available in the literature with which to compare the present study. Nevertheless, some of the results of the study conducted by Abdelhadi et al. (1998) on the effect of albumin stabilization on a soil's geotechnical properties may be useful for comparison. The Proctor dry density used in the manufacture of their specimens was 1.35 g/cm^3 and the specimens' aspect ratio was 2.2. In that work, the dry compressive strength increased with the addition of 1-2 wt % of albumin, especially when the specimens were oven dried at 50 °C (0.2 MPa to 0.8 MPa). Above all, the increase in the cohesion of the material (49 kPa to 111 kPa) was a macroscopic outcome that enabled the interaction between albumin and the soil to be understood. Regarding the other organic binders presented in the literature review, the maximum dry compressive strength was recorded as 7.0 MPa by Alhaik et al. (2017), who used 1 wt % of a particular starch on a fine clayey limestone soil.

Table 2 compares the compressive strength of the two soils when they were stabilized by ovalbumin, cement or hydrated lime. The data on the mineral binders were taken from Ouedraogo et al. (2020) as the specimen preparation and the tests were conducted in the same conditions as in the present study.

Table 2: Dry compressive strength (MPa) of the ovalbumin stabilized soils in comparison with cement and hydrated lime stabilized soils (Ouedraogo et al. 2020)

With the same amount of binder, ovalbumin improves the soils' dry compressive strength more than the mineral binders do. This is particularly interesting since mineral binders are generally assumed to be more effective than organic binders to improve soils' dry compressive strength and organic binders have mainly been used for improving soils' water resistance or for their low environmental impact compared to the mineral binders. Finally, the dry compressive strengths obtained for all the materials lie between 3 and 13 MPa and thus correspond to the classes (4 to 12 MPa) of (20cm*20cm*50cm) Hollow Concrete Blocks according to the standard (NF EN 15435/CN 2009).

4.1.2 Effect on the elastic modulus

Fig. 4a presents the stress-strain plots of the soils B and N when stabilized by ovalbumin or unstabilized. All plots have the same basic shape as illustrated in Fig. 4b.

Fig. 4: Stress-strain plots of unstabilized (a) and ovalbumin stabilized specimens of soils B and N (b)

Three zones can be distinguished. The "creep-like" zone A corresponds to the settlement of the specimen under load initiation. The larger this zone is, the looser is the specimen. Zone B represents the linear part of the material response once it becomes dense enough to show elastic behaviour under increasing load. Zone C is the post peak zone, which is related to the failure of the material. The flatter Zone C is, the more ductile is the material. Zone A exists for both unstabilized soils but it is significant on Soil N with 2 wt % and 4 wt % ovalbumin. In fact, drying cracks reported on those specimens induced additional porosity that was closed before the beginning of the elastic behaviour. For stabilized Soil B, since no drying cracks were found, the materials seem to have become firmer with ovalbumin addition. Regarding Zone C, stabilization makes the materials more brittle. Table 3 presents the elastic moduli calculated for the unstabilized soils and soils stabilized with ovalbumin and mineral binders.

Table 3: Calculated elastic modulus (in MPa) of Soils B and N when left unstabilized or stabilized with ovalbumin, cement or hydrated lime

Logically, the results for elastic modulus exhibit the same trend as those obtained for dry compressive strengths. The elastic modulus increases with the amount of binder. The most rigid materials are mixtures with ovalbumin. In addition, there is the effect of the nature and the density of the binding forces between the particles of the material (stabilized or not). The stronger and more numerous these bonds are, the less the material deforms. This would explain the difference between the types of binders. It can be seen that, regardless of the nature of the soil, the elastic modulus increases more with ovalbumin than with cement or lime. In the case of ovalbumin, adsorption forces and their high number on the surface of the soil particles due to the long chains of ovalbumin molecules, may explain the significant improvement in elastic modulus. There is a form of cross-linking of the soil matrix by ovalbumin molecule chains. Thus, an analogy can be made between the microstructure of soils stabilized by organic biopolymers and mother-of-pearl (Anger et al. 2013). Mother-of-pearl is a coating of the inner wall of certain mollusc shells. It consists of aragonite platelets ($CaCO_3$) interwoven by chains of conchyolin, which is a chitinoidal scleroprotein.

The method of determining the elastic modulus values discussed in this section are not very accurate and are therefore used for comparison. Cyclic loading and LVDT sensors were more suitable for accurate measurement of elastic moduli. Nevertheless, for most authors, the values of the elastic moduli of stabilized earth or raw earth without fibres are of the order of 200 MPa to 1000 MPa (Quagliarini et al. 2010; Bui et al. 2014; Miccoli et al. 2015) a range that includes the values found in the present study.

4.1.3 Compressive strength after two hours of wetting

Table 4 presents the wet 2 hours compressive strengths of the various mixtures of the two soils with 2 wt % and 4 wt % ovalbumin and also cement and hydrated lime.

Table 4: Compressive strength of the unstabilized soils B and N and corresponding ovalbumin, cement and hydrated lime stabilized soils after two hours of wetting

As was to be expected, the unstabilized soil specimens did not withstand the 2 hours' immersion in water. Therefore, no compressive strength was recorded for them. However, as for the mechanical strengths in the dry state, mixtures with 2 wt % and 4 wt % ovalbumin showed the best water resistance. In fact, with 4 wt % addition, the wet compressive strength of Soil B was 2.6 MPa, compared with 0 MPa at 2 wt %, while the same mixtures with soil N gave 2.9 MPa and 5.7 MPa. These values far exceed those obtained with mineral binders. Here, the water repellent property of the ovalbumin gives the reason for that improvement: the amphiphilic molecules fix themselves on clayey soil particles by their hydrophilic poles and form a water barrier with their hydrophobic poles once the matrix hardens. Ovalbumin is observed to be more effective on the water resistance of soil N than on that of soil B. This could originate from stronger interactions between Soil N clayey particles and ovalbumin molecules. Similar results were not found in the literature when considering the use of natural organic binders for soil stabilization. The use of cooking oils mixed with quick lime (Camões et al. 2012) or cow dung (Yalley et al. 2013) barely improved the soil's wet compressive strength compared to the results of the present study.

For the water resistance, it is difficult to compare stabilized soil specimens with hollow concrete blocks. It is well known that the resistance of the latter to water is "infinite", whereas the specimens under study resisted under particular test conditions that did not allow conclusions to be drawn on what would happen if the specimens were immersed for much longer periods of time. Nevertheless, it can be argued that raw earth bricks (even if stabilized) should not be used for applications where they would be immersed for a long time. In any case, the problem of the sensitivity of stabilized clay bricks remains and there is a significant reduction in compressive strength after immersion, which is not the case for hollow concrete blocks.

For building construction, Danso et al. (2015) refer to the Turkish standard (TS 704 1983, 1985), which recommends a minimum strength of 1 MPa, while Houben et al. (2006) suggest a minimum of 2 MPa for cement-stabilized blocks. According to the criteria of these two research teams, all materials in a dry state in the present study are suitable for construction. After 2 hours in a wet state, soils B and N stabilized with 4 wt % ovalbumin and soil N stabilized with 2 wt % and 4 wt % ovalbumin satisfied both criteria. Based on their compressive strengths, these materials are appropriate for housing construction.

4.2 Hygrothermal performances

4.2.1 Effect on thermal conductivity

Table 5 presents the thermal conductivities of stabilized soils, measured by the Guarded Hot Plate (GHP) and Hot Wire (HW) methods for the mixtures with ovalbumin, and by the GHP method for mixtures with mineral binders. The specimens were oven dried at 50 °C and cooled down in a super drier before the test. They were tested at the age of 45 days.

Table 5: Thermal conductivities of the unstabilized and ovalbumin-, cement- and hydrated lime-stabilized soils B and N

The thermal conductivity values measured by the hot-wire method were 0.15 W/(K.m) higher than those given by the guarded hot-plate method. This difference is well documented in the literature (Seng et al., 2019). The hot-wire method is easier and faster to implement but is less accurate because it is local and depends on the quality of contact between the probe and the surfaces of the two samples of material. The guarded hot plate method values are therefore considered in the following discussion.

There was no significant difference in thermal conductivity between the mixtures since all values measured were between 0.49 and 0.61 W/(K.m), whatever the different binders or different soils. This was due to the fact that all specimens were manufactured at very close dry bulk densities - all between 1.86 and 1.88 g/cm^3 - and thermal conductivity is strongly correlated to the dry bulk densities of materials (Laborel-Préneron et al., 2018; Liuzzi et al. 2013).

The usual thermal resistance of hollow concrete blocks with dimensions 20 cm x 20 cm x 50 cm is 0.23 m^2 K/W. For a thickness of 20 cm, stabilized earth bricks would have a thermal resistance of between 0.33 and 0.41 m^2.K/W, which is slightly higher than the thermal resistance of hollow concrete blocks. However, as the difference in thermal resistance is very small, it can be concluded that, for this property, the two materials are equivalent.

4.2.2 Effect on the MBV

Fig. 5 presents the moisture buffer values of Soils N and B according to the ovalbumin stabilization.

Fig. 5: The moisture buffer capacity of unstabilized and ovalbumin stabilized N and B soils with the Nordtest classification (Rode et al., 2005)

It is worth noting that Soil N is more hygroscopic than Soil B. This is due to the different nature of the clay minerals contained in the two soils. In addition to illite, Soil N contains chlorite and montmorillonite, which have larger specific surface areas and water absorption capacities than the kaolinite contained in Soil B. Similar observations have already been made on the same type of soils during water vapour sorption-desorption measurements (Cagnon et al. 2014; McGregor et al. 2014). For both soils, the addition of ovalbumin decreases the hygroscopic performance of the material even more than the mineral binders (2.40-2.80 g/(m^2.%RH) determined by Ouedraogo et al. (2020)). This clear difference is a consequence of the surface-active properties of the ovalbumin, which has prevented the materials from water degradation so far. Ovalbumin molecules adsorb to the -OH sites of the clay particles thanks to their hydrophilic poles, and their hydrophobic poles form a water-repellent film on their surface. This mechanism, which prevents the penetration of liquid water into the material, also limits the attachment of water molecules from the air to its intraporous walls during hygroscopic exchanges with the surrounding environment. However, up to 4 wt % addition, the hygroscopic performance of the materials treated with ovalbumin remains good according to the Nordtest criteria (Rode et al. 2005) in the case of moderate stabilization.

For comparison, the best values given by Rode et al. (2005) for lightweight aggregate concrete blocks, aerated concrete blocks and concrete barely exceed 1 g/(m^2.%RH), against more than 1.5 g/(m^2.%RH) in the present study. As expected, hygroscopic ability, which is the main quality of raw earth, remains better than that of hollow concrete blocks.

4.3 Effect of ovalbumin on soil microstructure

Microstructural analyses were performed in order to find the mechanisms underlying the macroscopic improvements observed with ovalbumin addition to soils.

4.3.1 XRD

Fig. 6 shows the XRD results for the Soils N and B when unstabilized or stabilized with 4 wt % ovalbumin.

Fig. 6: X-ray diffractograms of unstabilized soils (Soil N, Soil B) and stabilized soils with 4 wt % of ovalbumin (N4Ov and B4Ov)

By comparing the XRD of unstabilized and stabilized soils, no additional crystallized phase potentially formed by an interaction between ovalbumin and soil was found. A possible explanation could be that the small amounts of binder (≤4 wt %) did not permit meaningful phase formation that could be identified by XRD. Lagaly et al. (2006) and Chiu et al. (2014) reported that some organic polymers can intercalate between the interlayers of clay minerals, thus inducing an increase in clay mineral d-spacing, which can be shown by a left shift of the corresponding clay mineral peaks on an XRD pattern. However, the XRD analyses performed on the mixtures used here did not highlight this phenomenon.

4.3.2 Infrared analysis

Fig. 7 presents the infrared spectra of ovalbumin and of the soils N and B, unstabilized and stabilized with 4 wt % ovalbumin.

Fig. 7: Infrared spectra of unstabilized soils (Soil N, Soil B), soils stabilized with 4 wt % ovalbumin (N4Ov, B4Ov), and ovalbumin alone (Ov)

The ovalbumin spectrum shows peaks around 1400 cm^{-1}, 1545 cm^{-1}, 1640 cm^{-1}, 2970 cm^{-1} and 3300 cm^{-1}. According to Anger and Fontaine (2013), these bands correspond to the C-H bond twist (1480-1300 cm^{-1}), the C-N-H bond twist (1565-1500 cm^{-1}), the C=O double bond vibration (1750-1600 cm^{-1}), the C-H vibration (3100-2800 cm^{-1}) and the N-H bond vibration (3400-3200 cm^{-1}) characteristic of ovalbumin. The N4Ov and B4Ov spectra are almost identical to that of unstabilized soils. Yet, there are many interactions between ovalbumin and soils as indicated by the strong impact of ovalbumin observed on the mechanical and hygroscopic performance of the soils. This could be due to the characteristic bands of ovalbumin alone being hidden by those of the soils. Moreover, the 4 wt % ovalbumin quantity would be too low for its characteristic peaks to appear in the mixture. Nevertheless, weak energy bonds could be expected as the underlying reason for the materials' mechanical strength improvement, since ovalbumin contains carbonyl groups, amide groups and sulfoxide groups that are respectively reported as hydrogen bond acceptors, hydrogen bond donators and high-dipole moment groups (Lagaly et al. 2006). All these compounds are likely to establish low energy bonds with the clay minerals' interlayer hydroxyl groups (-OH) (Frost et al. 1998; Lagaly et al. 2006; Chiu et al. 2014).

4.3.3 SEM and EDX investigations of stabilization effect on the microstructure

Carbon coated pieces having polished and rough fracture surfaces were observed using SEM and EDS on samples used for dry compressive strength (soils N and B stabilized with 4 wt % ovalbumin). The observations were the same for both soils (N and B) so only the results obtained on soil N are presented on Fig. 8.

Fig. 8: SEM and EDS images of Soil N
(a, b) : unstabilized soil ; (c, e) : soil stabilized with 4 wt % ovalbumin (N4Ov)

The polished surface of the unstabilized Soil N appears to be firm, while cracks can be observed on the ovalbumin stabilized soil, confirming the microscopic cracks seen after manufacture of the specimens. But the cracks are filled with a substance showing a high sulfur concentration, whereas no sulfur was found in the unstabilized soil. On the fractured surfaces of ovalbumin stabilized samples, the zones showing sulfur concentrations were zoomed up to X500. This revealed the presence of a gel in these parts, which was formed by the ovalbumin. Egg white is known to contain sulfur, which contributes to its hardening when heated in the range of 61-84°C (Mine 1995). In normal conditions of use, the buildings are not expected to support such high temperatures. All these observations lead to the assumption that ovalbumin has ambivalent effect on the soil. In the humid state, its surfactant properties may compete with the swelling of clays and induce drying cracks but, when the material dries, its gel seals the cracks and strongly glues the soil particles together thanks to its sulfoxide and amide groups. Furthermore, ovalbumin may not be the only protein to interact with the soil particles, as secondary egg white proteins have also been reported to have similar properties. According to Mine (1995) and Liu et al. (2019), ovotransferrin has metallic binding ability, especially with Fe, Al, Cu, and Zn. Finally, almost all the egg white proteins have some tensioactive ability.

5. Conclusion

Vernacular construction techniques around the world have inherited “folk remedies” that resort to natural organic binders for stabilizing unfired earth. These techniques have been basically assumed to be environmentally friendly. However, there is a need to assess their effectiveness and to apply them to find out whether they can lead to modern sustainable and durable constructions with earth.

This work was carried out with the intention of measuring the effectiveness and of understanding the mechanism of stabilizing unfired earth with a natural organic binder that is the main protein of chicken egg white (ovalbumin). Two types of soil having different mineralogical compositions were tested in order to allow the results obtained in this study to be generalized to numerous soils. Soil B consisted of illite, muscovite and kaolinite, while Soil N contained calcite, illite, chlorite and montmorillonite.

The dry compressive strength, the water resistance and the hygrothermal performances of such stabilization have been discussed and compared to cement and hydrated lime stabilization. The addition of 2 wt % and 4 wt % of ovalbumin increased the Soil B compressive strength by a factor of about 3 (and 2 for Soil N). These improvements exceed the effect of the same amount of cement and hydrated lime stabilization. The elastic modulus measurement also shows the same trend. The material stiffness increases with ovalbumin addition even more than with cement and lime. Regarding the water resistance, significant improvements were brought by the ovalbumin addition. The ovalbumin stabilized soils withstood two hours of immersion in water and still had compressive strengths above 2 MPa, except for Soil b with 2 wt % ovalbumin. Once again, these results are better than those for mineral binder.

The thermal conductivity did not change, regardless of the soil nature or the type of binder, since the specimens had the same dry density. Conversely, the moisture buffer value (hygroscopy) decreased more with stabilization by ovalbumin than with stabilization by mineral binders. However, the MBV of stabilized specimens remained at least “good” according to the Nordtest criterion.

The XRD and the infrared analyses did not fully elucidate the mechanisms of the ovalbumin interactions with the soils. Nevertheless, the SEM-EDS showed the gel formed by this binder within the microstructure of the soil, filling the microcracks by gluing the soil particles together. It was also found that sulfur mapping could help to highlight the presence of ovalbumin in unfired earth materials.

All these outputs confirm that some natural organic binders, e.g. chicken egg white, can provide more effective stabilization of unfired earth than mineral binders do. However, further works could assess other durability parameters, such as the resistance to abrasion, surface erosion and fire; the risk of fungal growth; and the chemical durability of ovalbumin.

Ultimately, discussions could concern the practical use of such a natural organic binder considering its availability and the related ethical questions. This study was intended only to demonstrate the scientific effectiveness of using natural organic binder taken from old and vernacular techniques as an alternative to cement or lime. This proof of concept could encourage upcoming studies to focus on reducing the amount to that strictly necessary, given the ethical and economic issues, or on exporting the mechanism to organic binders from plants or synthetic molecules having comparable properties to ovalbumin and that could be more available.

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Caption for figures

Fig. 1: Mechanical measurements (a): Compressive strength; (a,b): Elastic modulus

Fig. 2: Compressive strengths of the ovalbumin stabilized soils B and N

Fig. 3: Occurrence of drying cracks on specimen of soil N stabilized with ovalbumin

Fig. 4: Stress-strain plots of unstabilized (a) and ovalbumin stabilized specimens of soils B and N (b)

Fig. 5: The moisture buffer capacity of unstabilized and ovalbumin stabilized N and B soils with the Nordtest classification (Rode et al., 2005)

Fig. 6: X-ray diffractograms of unstabilized soils (Soil N, Soil B) and stabilized soils with 4 wt % of ovalbumin (N4Ov and B4Ov)

Fig. 7: Infrared spectra of unstabilized soils (Soil N, Soil B), soils stabilized with 4 wt % ovalbumin (N4Ov, B4Ov), and ovalbumin alone (Ov)

Fig. 8: SEM and EDS images of Soil N (a, b) : unstabilized soil ; (c, e) : soil stabilized with 4 wt % ovalbumin (N4Ov)

Caption for supplementary materials

Table 2: Dry compressive strength (MPa) of the ovalbumin stabilized soils in comparison with cement and hydrated lime stabilized soils (Ouedraogo et al. 2020)

Table 3: Calculated elastic modulus (in MPa) of Soils B and N when left unstabilized or stabilized with ovalbumin, cement or hydrated lime

Table 5: Thermal conductivities of the unstabilized and ovalbumin-, cement- and hydrated lime-stabilized soils B and N

Table 1: Geotechnical, chemical and mineralogical characteristics of the raw soils B and N (Ouedraogo et al. 2020)

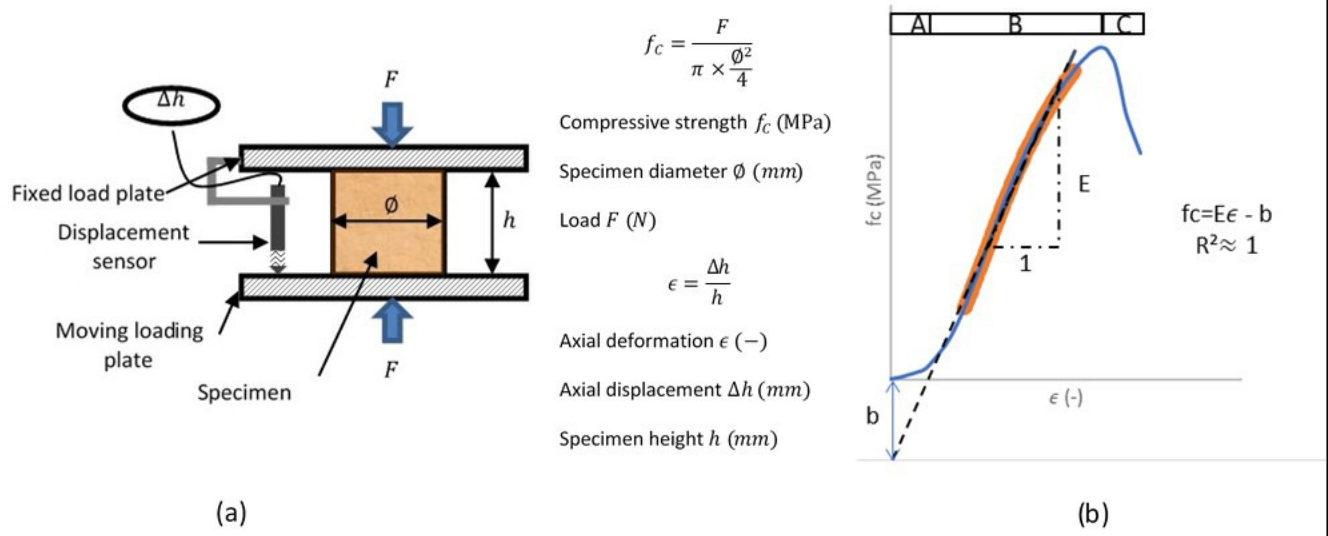
Geotechnical characteristics												
	Clay <2µm (wt %)	Silt 2-63µm (wt %)		Sand 63-2000µm (wt %)		Gravel >2mm (wt %)	w_l (wt %)	I_p (wt %)	Methylene Blue Value (g/100g)		w_{NPO*} (wt %)	ρ_{NPO*} (kg/m^3)
Soil N	22.5	38.5		37.0		2.0	46	15	4.10		14.1	1880
Soil B	32.0	40.0		28.0		0.0	38	21	2.65		15.6	1860
Chemical composition												
	SiO_2	Al_2O_3	Fe_2O_3	MnO	MgO	CaO	Na_2O	K_2O	TiO_2	P_2O_5	LOI	
Soil N	55.47	14.05	4.71	0.06	2.29	7.31	0.66	2.96	0.54	0.16	11.78	
Soil B	61.61	17.21	5.58	0.07	1.34	1.87	0.16	3.83	0.73	0.12	7.51	
Mineralogical relative composition												
	Q**	F**		I**		Mu**	C**	G**	K**		Mo**	Ch**
Soil N	++	+		++		+	++	+			++	+
Soil B	++	++		+		++	+	+	++			

**NPO (Normal Proctor Optimum)*

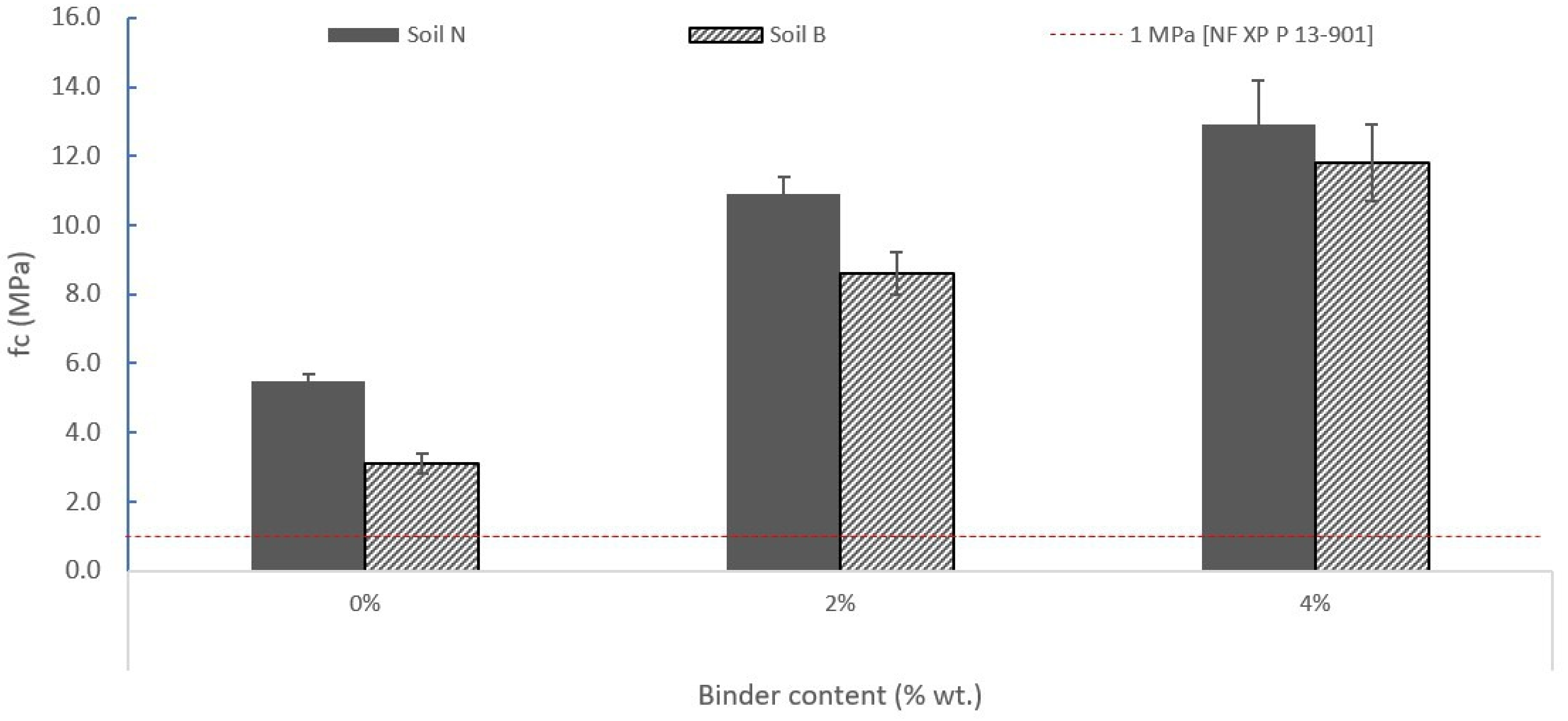
*** Q (quartz), F (feldspars), I (illite), Mu (muscovite), C (calcite), G (goethite), K (kaolinite), Mo (montmorillonite), Ch (chlorite)*

Table 4: Compressive strength of the unstabilized soils B and N and corresponding ovalbumin, cement and hydrated lime stabilized soils after two hours of wetting

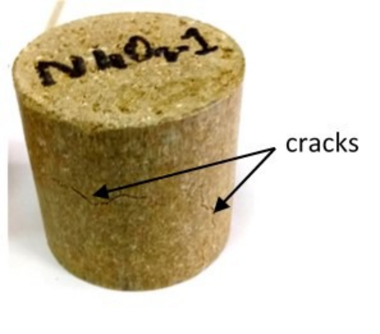
			wet fc (MPa)		
Soil	Binder (wt %)	ρ_d (g/cm^3)	Ovalbumin	Cement	Hydrated lime
Soil N	0	1.88	crumbled		
	2		2.9 ± 0.5	0.6 ± 0.1	0.6 ± 0.0
	4		5.7 ± 0.2	2.3 ± 0.1	1.0 ± 0.1
Soil B	0	1.86	crumbled		
	2		crumbled	crumbled	crumbled
	4		2.6 ± 0.3	0.6 ± 0.2	0.2 ± 0.0

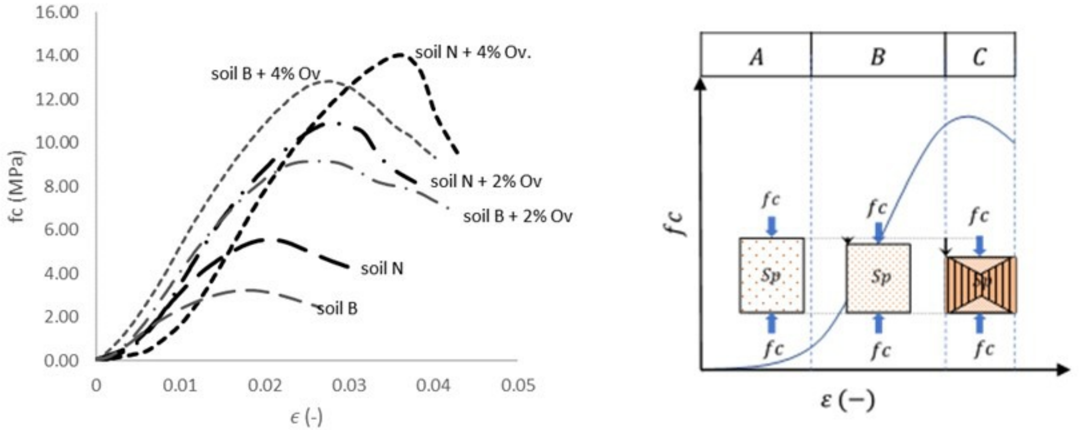


$f_C = \frac{F}{\pi \times \frac{Ø^2}{4}}$
Compressive strength f_C (MPa)
Specimen diameter $Ø$ (mm)
Load F (N)
$\epsilon = \frac{\Delta h}{h}$
Axial deformation ϵ $(-)$
Axial displacement Δh (mm)
Specimen height h (mm)
Δh
F
Fixed load plate
Displacement sensor
Moving loading plate
Specimen
$Ø$
h
(a)
A
B
C
fc (MPa)
E
1
fc=Eϵ - b
$R^2 \approx 1$
ϵ (-)
b
(b)

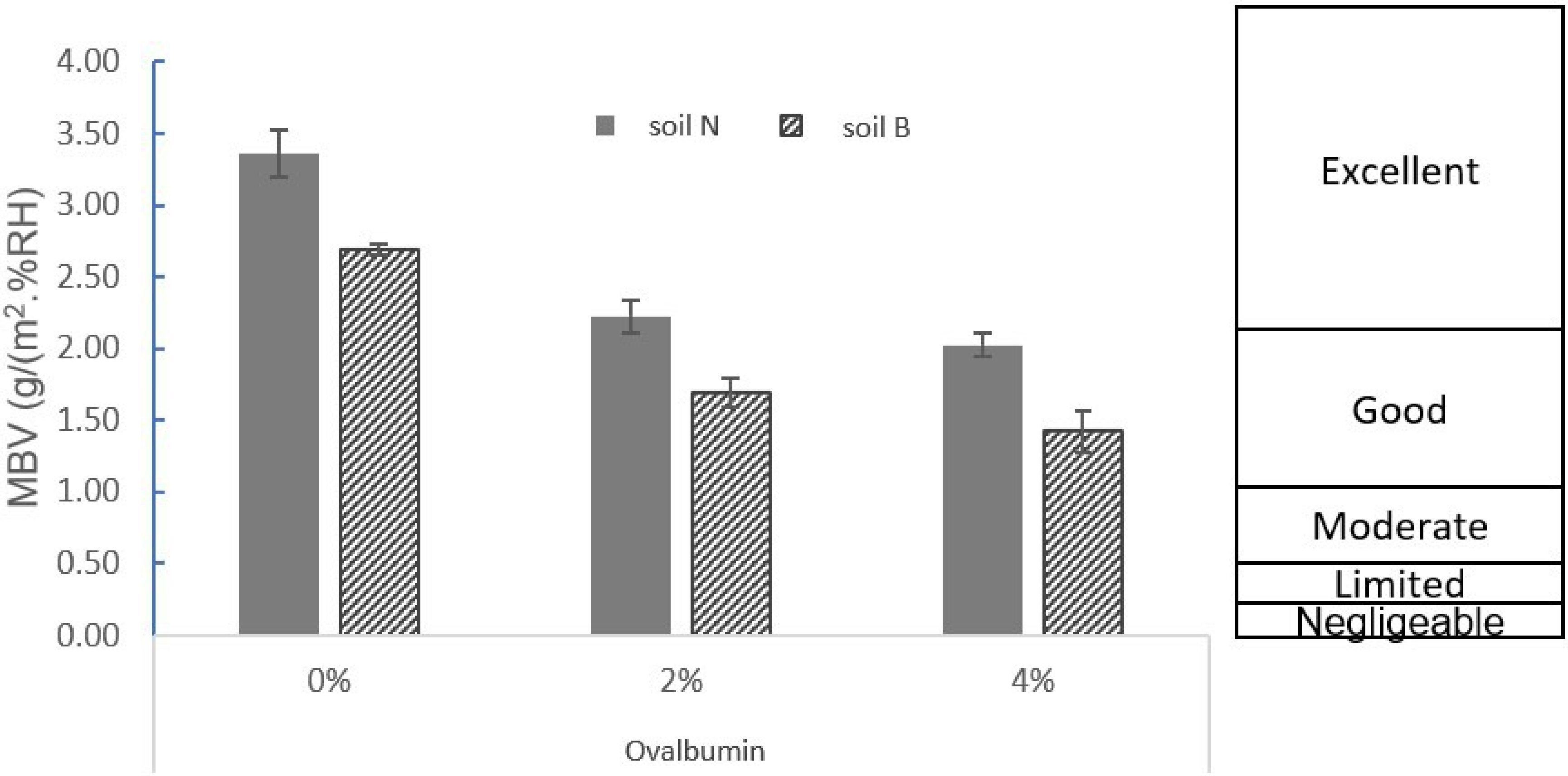


Soil N
Soil B
1 MPa [NF XP P 13-901]
16.0
14.0
12.0
10.0
8.0
6.0
4.0
2.0
0.0
fc (MPa)
0%
2%
4%
Binder content (% wt.)

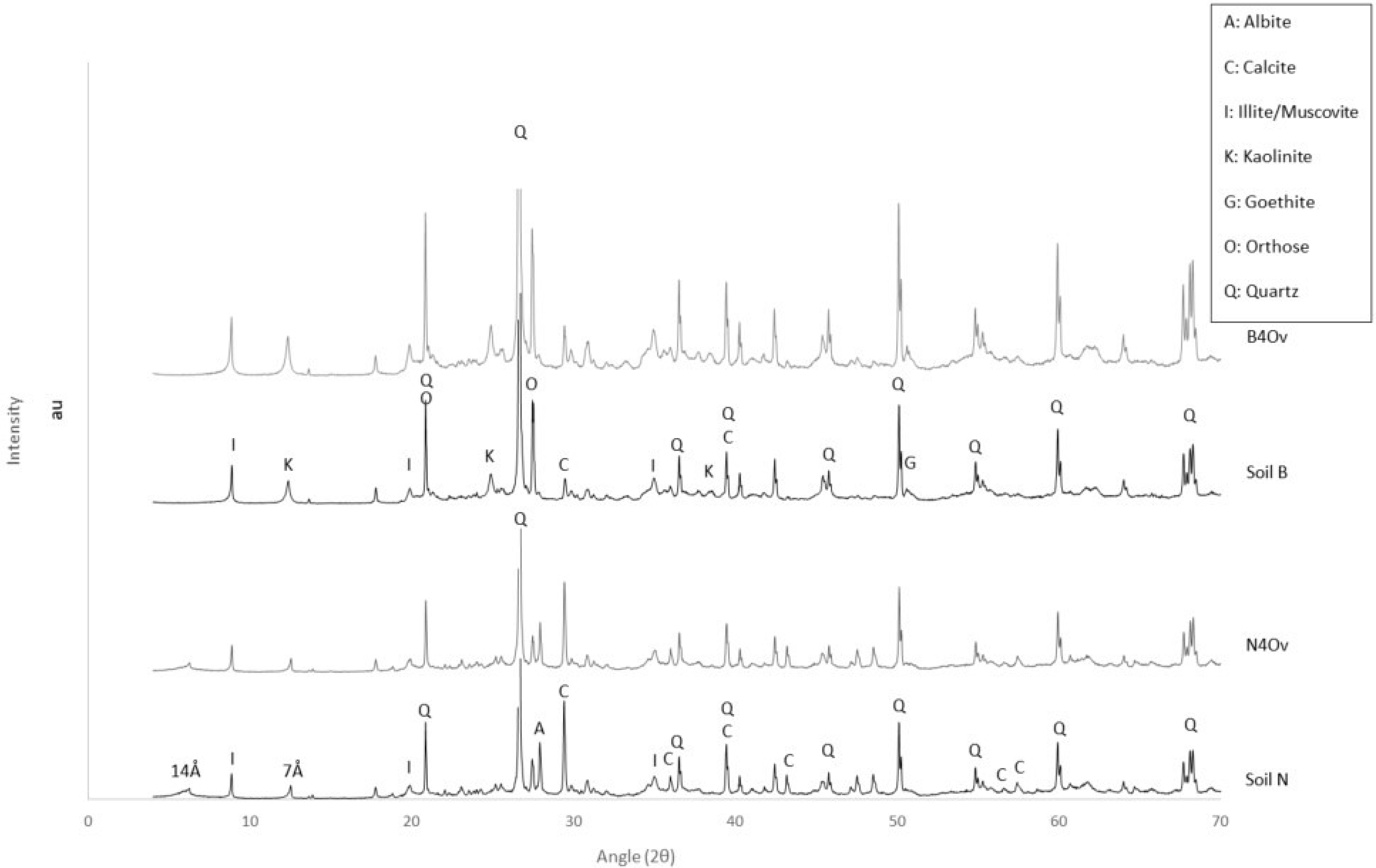

NH0v1
cracks


16.00
14.00
12.00
10.00
8.00
6.00
4.00
2.00
0.00
fc (MPa)
0
0.01
0.02
0.03
0.04
0.05
ϵ (-)
soil N + 4% Ov.
soil B + 4% Ov
soil N + 2% Ov
soil B + 2% Ov
soil N
soil B
A
B
C
fc
Sp
ε (−)

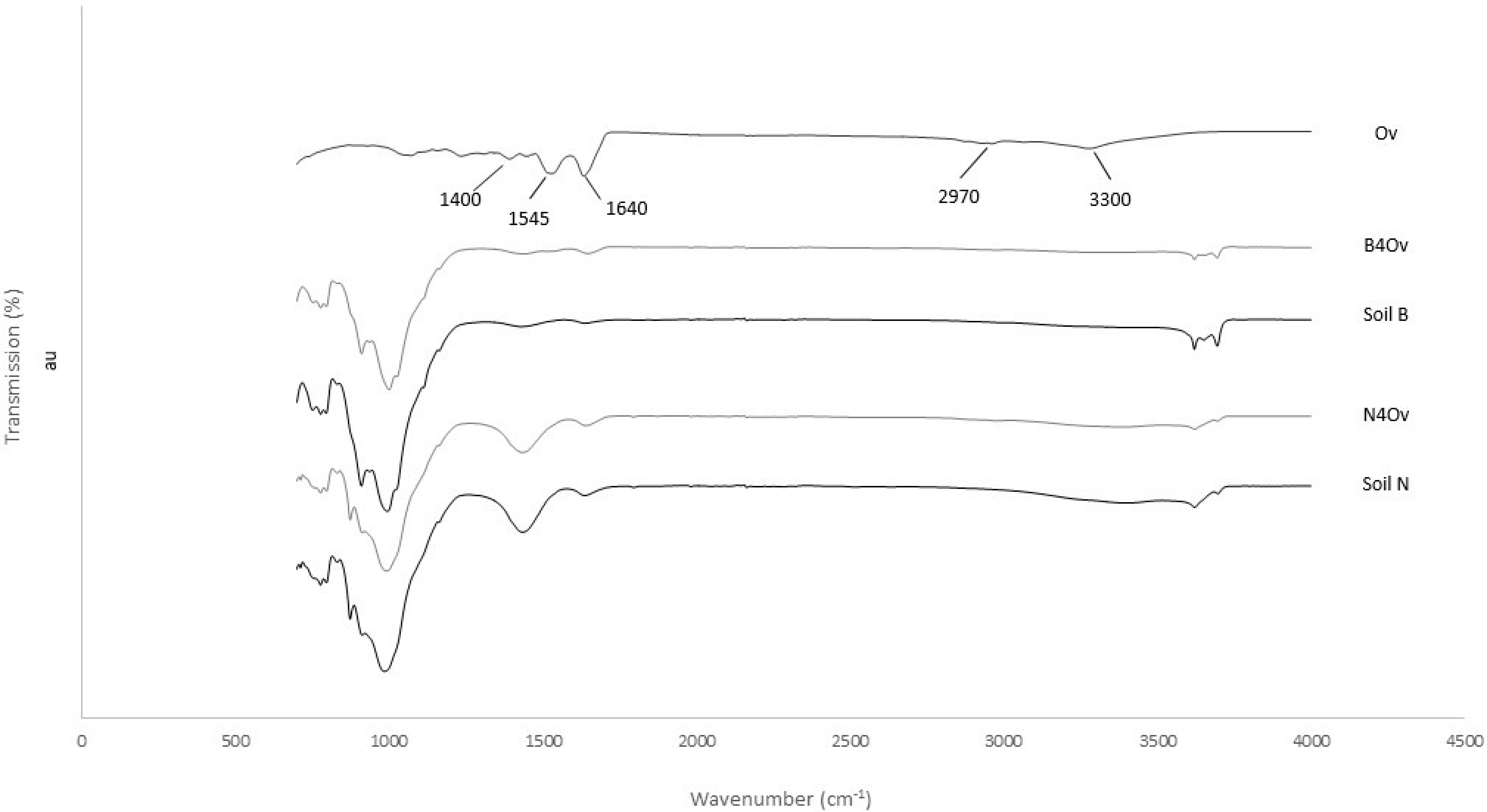
(a) (b)



4.00
3.50
3.00
2.50
2.00
1.50
1.00
0.50
0.00
MBV (g/(m².%RH)
soil N
soil B
0%
2%
4%
Ovalbumin
Excellent
Good
Moderate
Limited
Negligeable



A: Albite
C: Calcite
I: Illite/Muscovite
K: Kaolinite
G: Goethite
O: Orthose
Q: Quartz
Intensity
au
B4Ov
Soil B
N4Ov
Soil N
14Å
7Å
0
10
20
30
40
50
60
70
Angle (2θ)



Ov
B4Ov
Soil B
N4Ov
Soil N
1400
1545
1640
2970
3300
Transmission (%)
au
0
500
1000
1500
2000
2500
3000
3500
4000
4500
Wavenumber (cm-1)

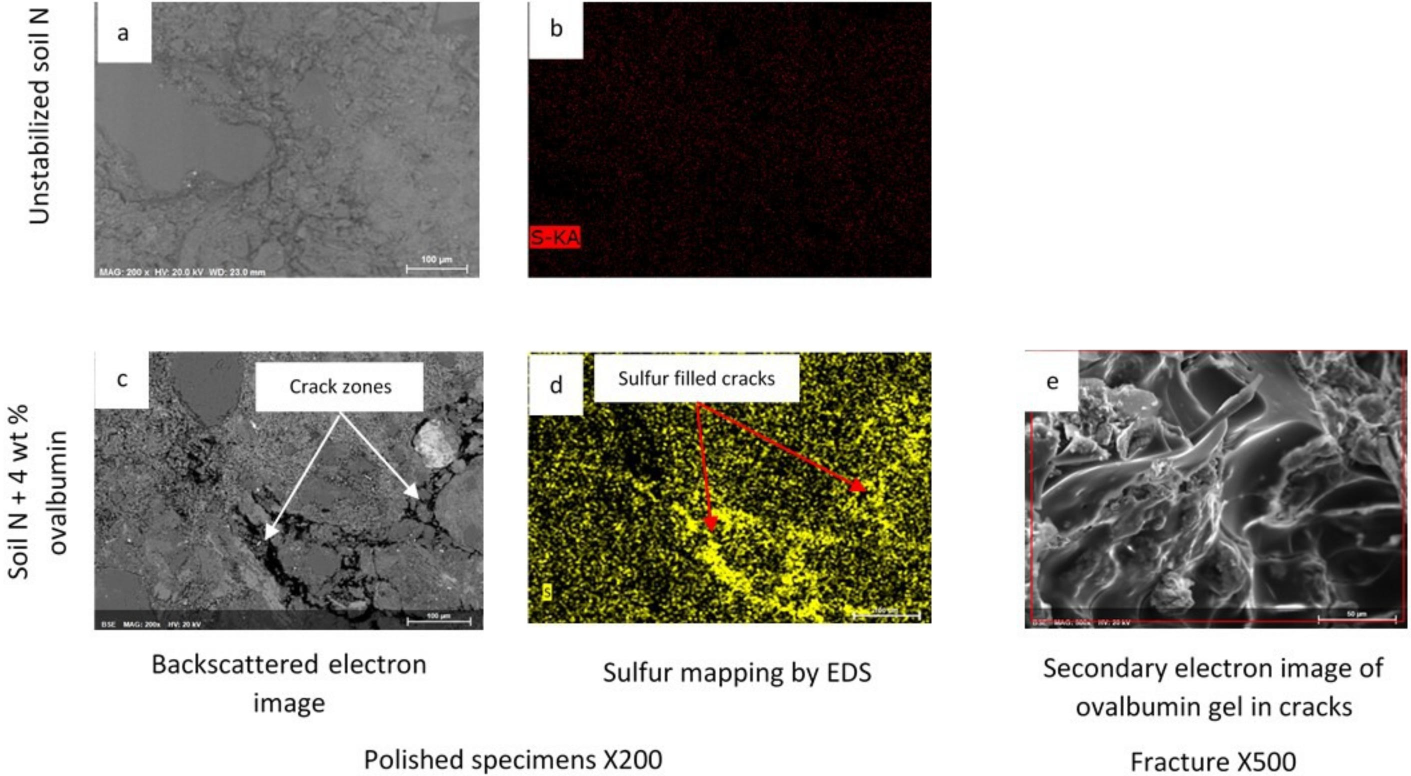

Unstabilized soil N
Soil N + 4 wt % ovalbumin
a
b
c
d
e
100 µm
S-KA
Crack zones
Sulfur filled cracks
Backscattered electron image
Sulfur mapping by EDS
Secondary electron image of ovalbumin gel in cracks
Polished specimens X200
Fracture X500

Table 2: Dry compressive strength (MPa) of the ovalbumin stabilized soils in comparison with cement and hydrated lime stabilized soils (Ouedraogo et al. 2020)

			fc (MPa)		
Soil	Binder (wt %)	ρ_d (g/cm^3)	Ovalbumin	Cement	Hydrated lime
Soil N	0	1.88	5.5 ± 0.2		
	2		10.9 ± 0.5	6.4 ± 0.3	5.5 ± 0.2
	4		12.9 ± 1.3	9.2 ± 0.2	5.3 ± 0.2
Soil B	0	1.86	3.1 ± 0.3		
	2		8.6 ± 0.6	4.8 ± 0.1	3.4 ± 0.1
	4		11.8 ± 1.1	5.8 ± 0.2	3.3 ± 0.1

Table 3: Calculated elastic modulus (in MPa) of Soils B and N when left unstabilized or stabilized with ovalbumin, cement or hydrated lime

			E (MPa)		
Soil	Binder (wt %)	ρ_d (g/cm^3)	Ovalbumin	Cement	Hydrated lime
Soil N	0	1.88	215 ± 18		
	2		352 ± 37	327 ± 14	262 ± 18
	4		525 ± 50	493 ± 18	319 ± 15
Soil B	0	1.86	203 ± 30		
	2		421 ± 40	265 ± 59	216 ± 20
	4		512 ± 40	275 ± 05	208 ± 16

Table 5: Thermal conductivities of the unstabilized and ovalbumin-, cement- and hydrated lime-stabilized soils B and N

		Ovalbumin		Cement	Hydrated lime
Soil	Binder (wt %)	GHP method ($W.K^{-1}.m^{-1}$)	HW method ($W.K^{-1}.m^{-1}$)	GHP method ($W.K^{-1}.m^{-1}$)	GHP method ($W.K^{-1}.m^{-1}$)
Soil N	0	0.56 ± 0.02	0.71 ± 0.03		
	2	0.60 ± 0.00	0.75 ± 0.02	0.53 ± 0.00	0.56 ± 0.02
	4	0.60 ± 0.04	0.78 ± 0.03	0.56 ± 0.02	0.56 ± 0.04
Soil B	0	0.60 ± 0.03	0.75 ± 0.02		
	2	0.61 ± 0.02	0.71 ± 0.01	0.49 ± 0.00	0.51 ± 0.02
	4	0.56 ± 0.03	0.70 ± 0.04	0.52 ± 0.02	0.50 ± 0.04