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Properties of unfired, illitic-clay bricks for sustainable construction

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HIGHLIGHTS

- Many of the world's top brick producers use illitic clay as a raw material.
- Unfired, illitic-clay bricks can be used in construction.
- The clay's properties are within limits suitable for earth construction, strengths fall within standard limits.
- An annual 15 million brick production, with half the units unfired, would economise over 4 million euros in kiln fuel and carbon tax in 10 years, and would approximately half emissions.

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ABSTRACT

Illitic clays are used for brickmaking worldwide. This paper explores the feasibility of using these clays unfired, to lower the environmental impact of construction. The results suggest that illitic clay can be used unfired. The geotechnical parameters of the clay are within the limits considered suitable for earth construction, and the compressive and flexural strengths fall within recommended limits in earth standards. Masonry constructed with the unfired illitic brick, bound with a standard hydraulic-lime mortar would reach a 28-day strength of 2.45 N/mm² which meets structural requirements in some European masonry standards.

Stabilization improved durability but lowered strength and vapour permeability, and did not significantly change thermal properties. The results suggest that both the quantity and composition of the clay fraction in an earth material determine the success of stabilization. The strength reduction caused by lime stabilization is mainly attributed to the adsorption of the Ca^{2+} by the illite, which prevents free Ca^{2+} from taking part in the pozzolanic reaction that builds up strength. The extremely high specific surface area of the illitic clay particles (24 m²/g) enhanced lime adsorption consequently damaging pozzolanic reaction and strength development.

If half of an annual production of 15 million brick was unfired, producers would economise over 4 million euros in kiln fuel and carbon tax in 10 years, and would approximately half their carbon emissions, lowering massively the global environmental impact of brick production.

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1. Introduction

As a result of pressing environmental problems, the construction industry (a large economic sector and one the greatest consumers of energy and natural resources) is adapting to the use of materials and methods of lower environmental impact. This paper promotes the use unfired clay brick to increase the sustainability of construction. It measures the properties of clay bricks, which are currently sold as a fired product, in an unfired form, and discusses

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their application and the environmental and economic benefit of their production.

Ceramic bricks are one of the most versatile building materials known. They have been used by Man since very early times and are still in use today, with a forecasted growth in years to come. IBIS reports a 7 billion dollar total revenue in 2019 worldwide, and forecasts annual growths over 2% for the next five years. Over 240 billion bricks are estimated to be produced annually in India (one of the main brick producers with China and Spain) which, with the building construction sector growing at a rate of 6.6% per year until 2030, will substantially increase demand in the next decades [1]. In the United States, about 27 million metric tons of clays were sold or used in 2018, valued at \$1.6 billion, and approximately half was common clay and shale for the production of brick

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and other clay products [2]. Brick manufacture has remained the main use for U.S. common clay and shale in the last years, as in 2015, it accounted for 41% of sales [3].

De Souza et al. [4], based on three different indicators including climate change, resource depletion and water withdrawal, state that ceramic brick walls have a lower environmental impact than concrete brick and reinforced concrete walls. However, firing produces greenhouse gases and consumes significant amounts of fossil fuel. The ceramic industry is energy intensive, as part of the process involves drying and firing to temperatures between 800 and 2000 °C. Furthermore, most industrial brick producers use unrenewable fossil fuel to fire their kilns and, as a result, they pay high annual taxes due to carbon dioxide emissions. Today, natural gas, liquefied petroleum gas (propane and butane) and fuel oil are mainly used for firing [5]. Due to the fuel combustion and the common presence of organic matter, sulphur and carbonate in the raw clay, gases such as carbon dioxide (CO_2) , sulfur dioxide (SO_2) , carbon monoxide (CO) and nitrogen oxides (NOx), are emitted on firing bricks, contributing to the world's greenhouse gas emissions. The economic growth forecasted for countries such as India and China in the next decades will carry a significant environmental impact. CO₂eq. emissions from the Indian brick kiln sector have been estimated by different sources at between 78 and over 200 million tonnes per vear [6].

In the context of sustainable building, increasing attention is given to the use of local materials and vernacular construction, in particular to earth construction. The sustainability of earth construction has been highlighted by previous authors. The environmental credentials of building with earth are obvious, as it involves using natural materials that can be sourced locally, have little or no industrial processing or preparation, and require modest energy for extraction. The manufacture of unfired clay bricks releases 80% less CO₂ into the atmosphere than fired clay bricks [7]. Furthermore, the end-of-life impact of earth-based materials is much lower than the impact of conventional building materials since they can be recycled with minimum effort and energy use.

Fernandes et al. [8] assess the life cycle performance and environmental impact of two earth materials (compressed earth blocks –CEB- and rammed earth- RE-), and compare them with ceramic brick and concrete block. According to these authors, 1 m² wall of RE and CEB account for half of the carbon emissions and embodied energy-EE- than ceramic brick or concrete block. They conclude that CEBs have an EE of 3.94 MJ/block (165 MJ per m² of wall), and a Global Warming Potential (GWP) of 0.39 kg CO₂ eq./block. However, in contrast, the environmental impact of fired bricks and concrete block are much higher, with EEs reaching 349 and 245 MJ per 1 m² of wall respectively, and GWPs of 57.4 and 82.6 kg CO₂ eq. respectively.

Venkatarama Reddy et al. [9], using a three-storey, load-bearing school building complex of a built-up area of 1691.3 m² as a case study, indicate that the EE of the earth building (1.15 GJ/m^2) is considerably lower than that of the fired clay brick building $(3-4 \text{ GJ/m}^2)$ and the reinforced concrete building $(4-10 \text{ GJ/m}^2)$. Similarly, Henry et al. [10], compare two typical residential buildings in Cameroon, one made of cement-blocks and the other with local mud-bricks. The authors conclude that the cement-block house has over 1.5 times more EE and emits at least 1.7 times more CO₂ than the equivalent mud-brick house.

In particular, unfired bricks such as CEB and adobe, constitute a valuable alternative material for sustainable construction. Christoforou et al. [11] studied the life cycle assessment –LCA- of adobe brick production based on the ISO standards and GaBi software. They compared the EE of adobe bricks with other materials in databases such as ICE (The Inventory of Carbon and Energy database) and previous studies to conclude that the EE of adobe bricks (0.03-0.17 MJ/kg) is much lower, than fired clay bricks (1-4 MJ/kg) and concrete blocks (12.5 MJ/block; 0.9 MJ/kg), even when transportation of the end-product to the building site is taken into account; and that the EE of adobe bricks is lower than the EE of other earthen building materials, such as stabilized rammed earth $(0.45-0.60 \text{ GJ/m}^3)$.

This paper concentrates on illitic clay. Illite is one of the most abundant clay minerals in sediments and rocks, and the major constituent of many brickmaking clays all over the world. The illitic clay studied comes from Kingscourt brick in County Cavan, Ireland. It is extruded, wire-cut and fired in tunnel kilns at 1030 °C to provide a durable, red brick of attractive appearance and a varied surface finish that has been used for building for several decades. This paper explores the possibility of using these bricks in an unfired form. First, the suitability of the illitic clay for earth construction is assessed by testing soil properties and comparing the results with the limits, reference values and recommended values considered suitable for earth construction. Then, the clay is mixed with sand and tested both raw and stabilized with hydraulic and hydrated limes. Several mixes are designed and produced in the laboratory and their physical properties tested to assess their mechanical, hygric and thermal performance. Finally the properties of the unfired material are compared with the fired brick, and the benefits of production discussed.

Earth as a construction material may not meet requirements for certain applications. Studies have shown that earth stabilization improves durability, strength, stiffness, workability and water absorption and load-deformation characteristics [12–15]. Therefore, the properties of the illitic clay were tested both stabilized and raw. Former authors propose that pozzolanic reaction between earth materials and lime results in cementing phases (calciumsilicate-hydrate and calcium-aluminate-hydrate) that enhance strength [13,15–17]. However, there are conflicting reports on the effect of stabilization on properties such as durability and mechanical strength, and the mechanisms taking place between the stabilisers and the earths are often unclear. Furthermore, the GWP and EE would increase significantly when a stabilizer such as lime or cement is added. According to Van Damme and Houben [18] the GWP of unstabilized raw earth would rise from 0.023 kgeqCO₂/kg to between 0.064 and 0.106 kg-eqCO₂/kg when stabilized with 5–10% of Portland cement –PC– (GWP of PC \sim 0.830 kg-eq. CO_2/kg).

2. Materials and methods

2.1. Properties and composition of the illitic clay

A Tertiary (Triassic Keuper), illitic clay quarried at the Cobaun and Cormey quarries at Kingscourt in County Cavan, Ireland, is studied. The clay in this paper is characterized closely so that the research results can be extended to other clays of similar properties and composition. The specific surface area was measured with the BET method which is based on the physical adsorption of gas molecules on a solid surface. The chemical composition was determined, as percentage by oxides, with an Epsilon 4 energy dispersive X-ray fluorescence (EDXRF) spectrometer. The mineral composition was analysed by X-Ray Diffraction (XRD), using the powder method, with diffractometer equipped with a PW1050/80 goniometer and a PW3313/20 Cu k-alpha anode tube at 40 kV and 20 mA. All measurements were taken from 3 to 60° (2θ) at a step size of 0.02° /second. The loss on ignition (LOI) was calculated, at 450 and 1000 °C, based on weight loss, to determine the carbon and carbonate content by thermal decomposition. The particle density was measured according with BS 1377-2 [19].

2.2. Mix design and preparation of test specimens

Initially, soil tests were undertaken on different clay and claylime mixes (Table 1) to determine the suitability of the raw clay for earth construction and the effect of lime stabilization. As the clay is predominantly silt graded (Table 2), sand was added to increase strength and lower linear shrinkage and moisture expansion. The sand's grading was determined by sieving in accordance with EN 933-1 [20] – Fig. 1. The sand contains only c. 6% fine gravel and a small range of particle sizes, with most grains sized between 0.1 and 2 mm and no clay fraction. The clay was stabilized with either natural hydraulic lime (NHL 3.5) or a hydrated lime (CL90S) complying with EN 459-1 [21] and manufactured by St. Astier and Clogrennane respectively. The lime is added as partial (clay + sand) replacement therefore the clay-lime mixes include slightly less sand than the clay specimens- Table 1. Compaction tests were carried out using variation of the Proctor's test, as explained below, to determine the optimal moisture content at (OMC) which the clay would achieve its maximum dry density by removal of air voids.

The materials were mixed with their optimum moisture content and prisms $(40 \times 40 \times 160 \text{ mm})$, circular discs $(100 \times 20 \text{ mm})$ and blocks (100x100 \times 100 mm) fabricated for testing. All the test specimens were prepared with a target bulk density of 2,200 Kg/ m³ by mixing a known mass in the specimen's cast volume. A high density was targeted, based on values in the literature and previous research by the authors, in order to optimise strength despite the likely increase of thermal conductivity. The specimens were cured for 28 days. First under wet hessian covered with a plastic sheet for one week. Later, the NHL3.5 specimens were placed in a wet chamber to complete curing at 90% ± 2% RH and 20 °C \pm 2 °C while the CL90 and clay specimens were cured at 60% ± 5% RH. Although lime hydration and carbonation might take longer than 28 days to complete, and hence strength could have improved if we had tested our specimens at 2 months or later, the 28-day target was chosen to be able to compare the results with most industrial and standard materials reported in the literature. Also, it is interesting to disseminate the standard 28-day strength because codes, lime and cement standards and site requirements are based on this value. The results are the arithmetic mean of at least three specimens, six for physical properties.

2.3. Soil properties: plastic and liquid limits (PL, LL), linear shrinkage (LS), plastic index (PI) and optimum moisture content (OMC)

The PL and LL were undertaken in accordance to EN ISO 17,892 [23]. The thread rolling method was used to determine the PL while the fall cone method was used for the LL. The linear drying shrinkage (LS) was measured in accordance to BS 1377 [19] and calculated from the equation in the standard. The clay was pre-

Table 2Grading of the clay as % by weight [22].

Fine gravel 2–5 mm	Sand 0.06–2 mm	Silt 0.06-0.002 mm	<0.002 mm	
0	1.5	83.2	15.3	



Fig. 1. Grading of the sand.

pared, placed in a shrinkage mould and dried in the air and in an oven.

The OMC was determined with a variation of the proctor test in BS 1377 [19], by uniformly compacting the mixes at different moisture contents in a cup of known weight and volume. The dry density of the mixes was determined according to the equations in the standard.

The dry density was then plotted against the corresponding moisture content from which the OMC and corresponding MDD were determined.

2.4. Physical property tests

The flexural and compressive strengths were measured in accordance to EN 1015 [24] – Fig. 2. The three-point loading test was undertaken to determine the flexural strength followed by the compressive strength test. The flexural and compressive strength were calculated according to the equations in the standards.

The water absorption coefficient due to capillary action ($C_{w,s}$) was measured in accordance with EN 1015 and involved sealing the prisms with paraffin leaving the base unsealed. The prisms were placed on their bases, in a tray with water to a depth of 5–10 mm and weighed at intervals of 1, 3, 5, 10, 15, 30, 60 and 90 min. The rate of absorption is calculated in accordance to EN 772 [25].

The water vapor resistance factor (μ) was measured with the dry cup test [26] which informs on the performance of materials,

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	Clay %	Sand %	Lime %	Tests	
				Soil properties	Physical properties
Clay + sand	50	50	0	OMC	Х
Clay + sand + CL90/5	47.5	47.5	5 CL90	OMC	Х
Clay + sand + CL90/10	45	45	10 CL90	OMC	Х
Clay + sand + NHL3.5/5	47.5	47.5	5 NHL3.5	OMC	Х
Clay + sand + NHL3.5/10	45	45	10 NHL3.5	OMC	Х
Clay	100	0	0	OMC, LL, PL, SL	-
Clay + CL90/5	95	0	5 CL90	LL, PL, SL	-
Clay + CL90/10	90	0	10 CL90	LL, PL, SL	-
Clay + NHL3.5/5	95	0	5 NHL3.5	LL, PL, SL	-
Clay + NHL3.5/10	90	0	10 NHL3.5	LL, PL, SL	-



Fig. 2. Device assembly for the three-point flexural strength test showing a clay specimen stabilized with hydrated lime.

at low humidity, when moisture transfer is dominated by vapor diffusion. The desiccant used was calcium chloride.

An adiabatic calorimeter was used to evaluate the specific heat capacity (SHC). Due to the anticipated disintegration of the unstabilized mix, the specimens were secured in tea bags to provide uniform testing conditions. The samples were heated and placed in the calorimeter in water of known temperature. The water temperature rise was monitored and the heat capacity determined according to the equation below:

$$c_m = \frac{m_l c_l (T_{li} - T_e)}{m_m (T_{mi} - T_e)}$$

where m₁-mass of water (g); m_m-mass of the specimen (g); c₁-specific heat of the liquid; T_{1i}-initial temperature of the liquid; T_{mi}-initial temperature of specimen (OC) and T_e-equilibrium temperature water/specimen.



Fig. 3. Casting of the thermal block with thermocouples placed at varying depths.

The thermal conductivity was measured with the guarded hot plate method in EN 12667 [27]. The thermal block (in which K type thermocouples had been placed in-situ at depths of 25 mm, 50 mm and 75 mm during casting- Fig. 3) was placed on top of a heating pad located on the guarded hot plate. With the power input maintained at 16 V and 0.7A, the variation in temperature at the thermocouple depths was automatically recorded for over 48 h until steady state conditions were achieved. Steady state conditions were considered to have been reached when the deviation in temperature was under 0.4 °C over a 4 h period. The thermal conductivity was then calculated using the equation below:

Thermal Conductivity,
$$\lambda = \frac{\emptyset \times d}{A(T_1 - T_2)}$$

where θ -average power supplied to the heating unit; T₁-average specimen's hot side temperature; T₂-average specimen's cold side temperature; A-metering area; d-average specimen thickness.

3. Results

3.1. Properties and composition of the illitic clay

The results evidenced that the material is an illitic clay including some carbonate (7.5% calcite- Ca CO₃, 5–7% LOI and 4% CaO)-Tables 3 and 4. The Ca²⁺ in the clay should contribute to the cation exchange and flocculation triggered by the lime reducing plasticity and increasing permeability.

According to the mineral analysis (Table 4), the clay includes clay minerals (57%) and non-clay minerals consisting of quartz (26%), calcite (7.5%), feldspar (4%), goethite (1%), hematite (4%) and rutile (0.5%); with the clay fraction consisting of illite (28%), chlorite (15%), smectite (5%), muscovite (4%) and fireclay (5%) (Table 4). Therefore, the illitic clay is slightly alkaline (7.5% CO₃Ca) which combined with the rising of the PH caused by the lime should enhance the dissolution of silica and alumina in the clay, and their combination with Ca²⁺ to form cementitious phases which enhance strength. Therefore, the alkalinity of the clay should contribute to hardening and strength enhancement. The availability of calcium in the clay may also enhance carbonation.

The specific surface area of the particles (SSA = 23.99 m²/g-Table 4) is extremely high, much superior to that of cement (CEM II /A-L 32, 5 N = 1.88 m²/g) and silicate materials used as pozzolans/supplementary cements such as FA and GGBS, with typical values of 6.5 and 2 m²/g respectively [29]. This high SSA should enhance the pozzolanic reactions between the clay (Si, Al) and the lime (Ca) to produce cementing hydrates that would strengthen the final product. The low LOI values at 450 °C (Table 4) will ensure that not swelling due to organic matter content will occur, and the LOI value at 1000 °C agrees with the calcium carbonate content determined by XRD and XRF. The clay meets the LOI requirements of Spanish standards UNE 41410 [30] for the fabrication of compressed earth blocks (organic matter <2%).

Table 4, shows the mineral transformations in the clay on firing and the mineral composition of the resultant brick. In the brick (1030 °C), the clay mineral (illite, chlorite/smectite) and calcite (CaCO₃) reflections have disappeared, and their Ca, Al and K have become part of new-formed, high-temperature aluminium silicates (potassium feldspar -<u>KAlSi₃O₈-</u> and gehlenite – Ca₂Al(AlSiO₇). These transformations agree with Peters and Iberg and Maggetti [31,32] who state that, in an illitic calcareous clay, illite-mica begins to transform at over 600 °C to disappear at 800 °C, chlorite disappears before 750 °C, and calcite decomposes at 750–850 °C with calcium silicates (gehlenite, diopside/wollastonite) and feldspar appearing in the 850 – 900 °C range.

Chemical composition by XRF - mean of the results in this paper and the producers results (**).

	SiO ₂	Al_2O_3	CaO	Fe_2O_3	Na ₂ O	K ₂ O	MgO	P_2O_5	SO_3	Cl	TiO ₂	MnO
Clay	55.84	19.76	4.92	8.71	0.00	5.63	3.05	0.60	0.04	0.04	1.17	0.12
Clay**	57–59	15–18	3.7-4.1	4.7-6.3	0.3-0.5	4.60	1.7-2.4	-	0.1-0.6	-	0.80	-
Arith mean	58	18	4	6	0.2	5	2.8	0.6	0.2	0.04	1	0.12

Table 4

Mineral composition of the clay and mineral changes on firing. Specific surface area, particle density and loss on ignition. *Lingl [28].

	Mineral composition				SSA m ² /g	Particle density Mg/m ³	LOI (%)	
	Major > 40%	Subsidiary 40–15%	Minor 15-7%	Traces < 7%			450 °C	1000 °C
Illitic clay (raw)	Total clay 57% *	quartz chlorite/smectite illite (muscovite)	calcite feldspar (Ca)	hematite	23.99	2.65	0.21 0.05*	5.10 6-7*
Illitic clay fired at 600 °C	quartz	feldspar (Ca)	calcite, muscovite/ilite		-	-	-	
Brick (illitic clay fired at 1030 °C)	quartz	feldspar (K)	gehlenite,	hematite	-	-	-	

3.2. Effect of lime stabilization on illitic clay properties

The liquid and plastic limits evidence the percentage of moisture at which a soil passes from the plastic to the liquid state and from the solid to the plastic state. Both limits depend upon the amount and type of clay present. A soil with high clay content usually has high liquid and plastic limits and a less cohesive (sandy or silty) soil lends low figures. As expected from the high clay content, the material subject to study shows high values.

The results (Table 5) evidenced that the lime increases both the plastic limit and liquid limit of the clay. However, the plasticity index is not affected (except for the 5% CL90 with a slightly lower PI). According to Cheshomi et al. [33], lime lowers the PI due to agglomeration caused by cation exchange taking place between the clay and the lime. The replacement of the monovalent ions (Na+ and K+) in the clay with the higher valence Ca^{2+} ions of the lime (cation exchange) reduces the electrical charge around the clay particles which come closer to each other forming flocs and causing the soil to agglomerate, becoming less plastic and more permeable [12,13,15]. However, in the illitic clay, the PI doesn't lower suggesting that cation exchange between the illitic clay and the limes is limited, and was probably enhanced using 5% hydrated lime (CL90s) stabilization. This might be due to the type of clay minerals present in the illitic clay. A PI reduction due to lime stabilization has been reported for kaolinite clay [34] suggesting an active cation exchange which is absent in the illitic clay studied. The type of clay mineral present heavily influences the plastic properties of lime stabilized soils [35], either increasing [12,36] or decreasing the PI [33,37].

The linear shrinkage of the clay alone is high, as expected from the high clay mineral content. As it can be seen from Table 5, the addition of lime lowers the shrinkage of the illitic clay. This agrees with previous authors [12,14,38,39]. However, this tendency is reversed at a certain lime content between 5 and 10%. Therefore, for the illitic clay, the optimum lime content for lowest shrinkage lies between 5 and 10%. Dash and Hussain [14] and Buhler and Cerato [40], also found this trend over a certain lime threshold. Based on the recommendations of maximum permissible linear shrinkage in Table 6, the 5% NHL3.5 mix is the best suited to meet requirements for earth construction.

3.3. Suitability of the illitic clay for earth construction based on the soil parameter results

As it can be seen from Tables 5 and 6, all the soil parameters of the illitic clay are within the limits considered suitable for earth construction. The low PI indicates that the clay will soften significantly with increasing moisture content which agrees with the significant clay content. According to Burroughs [40], based on an analysis of 219 specimens of RE, the PI and LS are the best discriminators of the soil's predisposition for a successful lime stabilization. The PI of the clay studied is well within the limits established by former authors, while the LS marginally exceeds some of the limits prescribed which is likely caused by the active clay content (chlorite/smectite and ilite). The PI vs PL values of the illitic clay meets the requirements of the Spanish standards [30] for the fabrication of compressed earth blocks; while some of the stabilized clay mixes (CL90 10%) are just outside the limit (Fig. 4).

The particle size distribution of earth materials is important and has been related to their strength and durability [7,15,52]. Sand was added to the illitic clay as explained in the methods section. According to the sand grading (c. 6% fine gravel and most grains sized 0.1–2 mm- Fig. 1) and the illitic clay (predominantly silt graded-Table 2), the material investigated lies within the granulometry values recommended for materials for earth construction (Table 7).

Table 5							
Soil parameters of the clay	with and without	stabilisers. PL-plast	ic limit; LL-liqu	id limit; PI-1	plasticity index;	LS-linear shrinka	ge.

Mix Composition	PL	LL	PI	Linear Shrinkage –LS %
100% clay	17.2	32.1	14.9	7
CL90 5%	24.4	37	12.6	3.9
CL90 10%	25.2	39.6	14.4	4.6
NHL3.5 5%	22.7	37.6	14.9	2
NHL3.5 10%	22.9	37.5	14.6	3.5

Comparison of the soil property values in this research with values for earth construction recommended by former authors. PL-plastic limit; LL-liquid limit; PI-plasticity index; LS-linear shrinkage.

Reference	PL	LL	PI	LS %	Allowable % LS
Tinsley & Pavia [41] Jimenez Delgado & Guerrero [42] Houben & Guillaud [43]	19 2-30	35 24-25 25-46: 25-51	17 2-28 2-30: 10-25	7.7 <2	<2.5 HB 195 [48] <0.05 NZS 4298 [49] 2 Keable and Keable [50]
Walker et al. [44] Walker [45]		<45	<2–30 5–15	<5	3 Little and Morton [51]
Burrows [40]			15; 15–30	<6; 6–11	
CDI & CRATerre [46] XP P 13-901 [47]	2.5–29 2.5–29	25–50 25–50			



Fig. 4. Plastic index vs liquid limit of the Illitic clay in relation with the recommended values for compressed earth blocs in Spanish standards [30].

3.4. Strength results: effect of lime stabilization and compliance with allowable earth construction values

The un-stabilized illitic clay reached the highest strength in both flexion (1.34 N/mm²) and compression (4.98 N/mm²)-Figs. 5 and 6. Therefore, lime stabilisation does not increase the strength of the clay-sand mix. Nevertheless, the strength of all the mixes falls within accepted/recommended limits for earth construction (\geq 1.3–2.1 N/mm² compressive strength in the codes and standards in Jimenez Delgado and Guerrero [42].

The flexural strengths (0.48–1.34 N/mm²) are within the recommended earth building guidelines in the USA, Australia and New Zealand. As a general guidance, the New Zealand standard [49] proposes a characteristic bending strength equal to 0.1 N/ mm² (when field test results are not available). More conservative, the Australian earth building handbook recommends ignoring any material strength in bending in the absence of test data [58].



Fig. 5. Flexural strength of the illitic clay with and without lime. KCC = clay.



Fig. 6. Compressive strength of the illitic clay with and without lime. KCC = clay.

According to ACI [59], the allowable flexural tensile stress or the modulus of rupture, for clay and concrete masonry is 0.206 N/mm² (30 psi), which is more than doubled by the unstabilised clay mix. The unstabilised clay mix and the NHL3.5-stabilised clay can be classified as class 3 and 5 according to Spanish standards [30].

The main reason of the strength drop on stabilisation is likely the high clay mineral content of the earth used in this research,

Table 7

Granulometry of the mixes tested compared with recommended values for earth construction (%) (*) in Jiménez Delgado and Guerrero [42].

Material/Source	Clay <0.002 mm	Silt 0.002–0.06 mm	Sand 0.06–2 mm	fine gravel 2-6 mm
Illitic clay + sand	8-9	40-42	45-47	3-5
Recommended granulometry for Rammed Earth				
SAZS 724 [53] *	5-15	15-30	30–75	
IETCC [54] *	10-40	20-40	10-40	10-20
McHenry [55] *	15	32	30	23
Recommended granulometry for Compressed Earth Block				
HB 195 [48] *	10-40	10-30	30-75	
Rigassi [56] *	8-30	10–25	25-80	0-40
Smith and Austin [57]*	4-15	40	60-80	-
Walker et al. [44]				
For RE and CEB, also PI = 2–22, LL < 40%	Up to 20	15 to 30	45 to 80	

as opposed to most earth materials in the literature which are usually richer in sand/gravel and silt. The abundant clay minerals in this earth adsorb the Ca^{2+} (lime fixation) which changes the plasticity of the material but does not contribute to strength development, since free Ca^{2+} is no longer available to take part in the pozzolanic reaction. The higher the lime fixation the lesser the Ca^{2+} available for pozzolanic reaction and strength increase. There is a direct correlation between the amount of lime fixed by an earth material (Lm) and its clay fraction (%) [60].

$L_{m} = (\% \text{ clay fraction } / 35) + 1.25$

When lime is added to an earth material, initially, all the available Ca ions are adsorbed onto the inter/intralayer surfaces of the clay minerals, due to their affinity which is owed to charge deficiency [60,61,62]. The adsorption of the Ca²⁺ by the clay (known as lime fixation - Lm), changes the plasticity of the earth (LL and PI) but does not contribute to strength development, since free Ca²⁺ is no longer available to take part in the pozzolanic reaction [60–62].

The strength drop on stabilisation can also be related to insufficient moisture available during curing. As aforementioned, the earth in this research (a brick-making mudrock) includes significantly more clay minerals than other earths in the literature. Clay minerals such as illite are eminently hygroscopic, they absorb water fast and are capable of retaining it. There is a competition for water between the clay minerals (initial imminent adsorption of water molecules onto their inter and intralayer surfaces), and the processes of lime hydration and carbonation which need moisture during curing to take place. The lack of sustained moisture during curing to complete these processed might have undermined strength development.

The slight reduction in the quantity of sand in the stabilized mixes may have contributed to the strength loss. Narloch et al. [63] claim that the gravel fraction increases the flexural strength of stabilized rammed earth and Guettala et al. [64] observe that the compressive strength of stabilized soil increases as the sand quantity grows from 0 to 40%.

Other authors have also found strength reduction on stabilisation. For each earth, there is an 'optimum lime content - OLC' that satisfies the immediate soil–lime interaction still providing sufficient free calcium necessary to initiate the pozzolanic reaction required to increase strength, hence, under the OLC, strength reductions can occur [60–62]. Minke [65] states that small percentages of cement stabilisation (<2%) resulted in a reduction in strength of a clay-rich soil and attributes this to the cement interfering with the inter-particle bonding of the clay. McPadden and Pavia [66] also found compressive strength loss in a glacial till stabilised with 5–15% lime, and blamed this on the high water demand of the soil's fine fraction undermining hydration and carbonation in the stabilized specimens. Temga et al. [67] also show that 4%-8% lime stabilization lowered the compressive strength of a clay including 25–75% sand.

Increasing NHL3.5 content from 5 to 10% increases both strengths however, increasing the hydrated lime content (from 5 to 10%) results in a strength reduction of c. 11%. Therefore, there is an optimum hydrated lime content beyond which further addition results in strength reduction agreeing with previous authors [14,17,60–62]. The compressive strength of the NHL3.5-stabilized mixes is clearly superior to the CL-90 mixes which concurs with Miqueleiz et al. [15] and is probably due to the strength contribution of the NHL hydrates. However, the flexural strength of the NHL3.5-stabilized mixes is either similar or lower than the CL-90 mixes.

3.5. Water absorption by capillary action

As expected, the un-stabilised clay and sand mix disintegrated when subject to the capillary test. However, all the stabilized, clay-sand mixes lasted well for the duration of the test. The rate of absorption lowers over time, and the CL90/5 mix registered the lowest absorption (628.5 g/min.m²), while the NHL3.5/10 reached the highest (995.17 g/min.m²)- Fig. 7.

In all cases, the water absorption increased when the lime content raised from 5% to 10%. This disagrees with former authors that found that increasing lime content in clay soil lowered water absorption [15,68]. The increase in water absorption with increasing lime content can be attributed to the lime being still active at the time of the testing: it is likely that the carbonation of the CL90s and the hydration of the NHL3.5 were not completed at the time of the testing (28 days). This would also explain the greater suction of the hydraulic lime specimens which require water to complete hydration. The greater imbibed amounts of water over a short time period are likely related to changes in the material pore size distribution due to particle flocculation. In total, the lime-stabilized mixes absorbed between 16 and 20% water with respect to their initial dry weight.

3.6. Water vapor permeability

Lime stabilization reduced the vapor permeability of the clay, hence increasing the water vapor resistance factor (Table 8); and the hydraulic lime mixes are more impermeable than the CL90 mixes. However, all the mixes comply with the German standard on earth bricks [69], that recommends water vapor diffusion factors in the range of 5–10 [70]. Therefore, the vapour permeability of the illitic clay, both raw and stabilized, are suitable for earth construction.

Furthermore, the vapor resistance factors are within the range of similar materials in former studies. The results are consistent or slightly lower than those of McGregor et al. [71] who observed that the addition of 4%-8% lime or cement stabilizer to clay resulted in a reduction in the vapor permeability- Table 9- due to crystal growth during lime hydration and carbonation reducing accessibility. According to Liuzzi et al. [72], the reduction in vapour permeability of lime stabilized clay is due to an increase in the specific surface area and the loss of mesopores causing tortuosity. The values are lower than the resistance factors of 7–8 reported for stabilized earth blocks made with laterite, sand and cement of density ranging from 1818-2000 kg/m³ [73]; and the 6–13 factors for densities 1600–2100 kg/m³ reported by [74].

3.7. Specific heat capacity

The specific heat capacity of a material indicates the material's ability to store heat. As expected, the specific heat capacity of the illitic clay/sand mixes is high, typical of high-density materials. The results show that the addition of lime to the clay/sand material increases the specific heat capacity. This is probably due to the



Fig. 7. Water absorption by capillary suction (g/min. m²) over time.

Permeability to water vapour of the illitic clay mixes compared with similar earth materials and conventional building materials. *WUFI PRO 2009- dry cup/wet cup. ** Walker and Pavia [75].

Table 9

Specific Heat Capacity of the illitic clay/sand mixes compared with other materials.

Source	Material	Specific Heat Capacity (J/ Kg.k)
Experimental	ILLITIC CLAY + SAND	945 ± 22
Results	CL90/5	1224 ± 102
	CL90/10	1134 ± 21
	NHL3.5/5	1065 ± 20
	NHL3.5/10	1198 ± 73
Soudani et al. [76]	RE 2.5% lime	648
AIRAH in Soebarto [77]	Standard value RE	1260
Adam et al. [78]	6% lime stabilized earth blocks	835-836
	5% cement stabilized earth blocks	835-836
Walker and Pavía	Lime plaster	863.9
[75]	Lime and hemp	1068.00
	Lime and cork	866.5
Tinsley and Pavia [41]	Glacial till + 5% lime	1218.66
Minke [79]	Loam soil	1000
Cagnon et al. [70]	Earth bricks	910-960
Houben et al. [80]	RE- highly compacted	1830

densification induced by the products of the reaction between the lime and the clay. The 5% CL90 stabilisation increased the heat capacity the furthest. However, there is no clear trend and the differences between the two limes are not significant. In order to evaluate the results, comparisons are drawn with former authors (Table 9). The values in the Table are slightly erratic as they reflect the variable nature of the materials and the influence of fluctuating compaction. However, the results obtained are in range with earth materials reported by previous authors, superior to lime-based materials and lower than highly compacted rammed earth.

3.8. Thermal conductivity

The thermal conductivity of the 5% CL90 (1.227 W/mK) may be slightly overestimated due to the 75 mm thermocouple being displaced during the preparation of the test specimen. With the exception of the clay/sand mix with 10%CL90, lime stabilization increases the thermal conductivity of the illitic clay. However, increasing the amount of lime does not seem to enhance the thermal conductivity. There are conflicting reports on the variation of thermal conductivity on stabilization. Some authors found that increasing the content of stabilizer increases thermal conductivity: Saidi et al. [81] noted that, when the lime content in earth bricks

was increased from 0 to 12%, the thermal conductivity increased from 0.7902 to 0.9785 W/(mK). In contrast, other authors show that increasing lime content (from 0 to 5%) lowered thermal conductivity by about 30% in three different soils [72]. The inconsistencies are probably related to the amount of reaction between the earths and the limes and the amount of phases (and densification) formed as a result. The thermal conductivity values of all the clay-sand mixes are within the standard ranges of some of the most common construction materials (Table 10 and Fig. 8).

4. Discussion

4.1. Effect of lime stabilization on the physical properties of the unfired, illitic clay brick

It was expected that the new phases produced by the lime's carbonation and hydration, and the (clay-lime) pozzolanic reaction would densify the structure increasing strength, specific heat capacity and thermal conductivity and lowering vapour permeability and capillary suction. The thermal conductivity, specific heat capacity and vapor permeability varied as expected. However, the strength of the clay lowered due to stabilization. The unstabilised mixes disintegrated during the capillary suction test, indicating that stabilization improved the durability of the illitic clay.

It was also expected that the hydrolysis of the abundant free lime in the system (the hydrated lime includes 90% Ca(OH)₂ and the NHL3.5 25% available lime) would issue abundant OH⁻, rising the PH and enhancing the dissolution of the clay's silica and alumina, and hence their combination with the lime's Ca²⁺ to form cements that would contribute to strength (pozzolanic reaction). The Ca²⁺ naturally occurring in the illitic clay (7.5% CO₃Ca) and the great specific surface area of the clay particles (superior to cement and silicate materials used as pozzolans) should have enhanced pozzolanic reactions as well as carbonation. The stabilized specimens were cured at high humidity to facilitate these reactions. However, the results suggest that reaction was very limited. As aforementioned in the strength results, this is attributed to a combination of three factors:

1-The adsorption of the Ca2 + by the clay minerals (lime fixation) which prevents free Ca2 + from taking part in the pozzolanic reaction leading to strength development; 2-the competition for water between the hygroscopic clay minerals in the illitic clay and the lime, 3- and the short curing (carbonation and pozzolanic reaction are slow and the tests took place at 28 days). The extremely high specific surface area of the clay particles (SSA = 23.99 m²/ g-Table 4), facilitated lime fixation in the inter and intralayer clay surfaces consequently damaging pozzolanic reaction and strength development.

Nevertheless, despite the limited reactions in the system, the compressive strength of all the mixes falls within accepted/recommended limits for earth construction, and the flexural strengths are



Fig. 8. Variation in thermal conductivity triggered by lime stabilization.

Thermal conductivity values of the clay-sand mixes compared with standard thermal conductivities of common construction materials in EN 12524 [82].

Material	Thermal conductivity (W/mK)
Concrete (1800 – 2200 Kg/m ³)	1.15 – 1.65
Lime and Sand Plaster	0.8
Gypsum and Sand Plaster	0.8
Cement and Sand Plater	1.0
Clay or Silt	1.5
Sand and Gravel	2.0

significantly superior to the allowable limits in USA, Australia, Spain and New Zealand standards.

In the hydraulic lime mixes, hydraulic reactions were expected to form cementing hydrates. The formation of a network of hydrates is often considered the reason for the increase in strength and thermal conductivity and the drop of water absorption. However, this has not yet been demonstrated, and with the relatively low volume fraction of cement/ hydraulic lime in stabilized earth, it seems unlikely that a network of hydrates could be reached [18].

The results indicate that stabilization of the illitic clay (at low lime contents) improves durability in liquid water, it lowers strength and water vapour permeability, and does not significantly change thermal properties. The lime makes the clay more impermeable to water vapour, but the water vapor diffusion factors are still within the values recommended by the German standard for earth bricks. Van Damme and Houben [18] state that stabilization is often unnecessary and has a limited role apart from helping the material to pass durability tests that are inadequate for earth materials. The authors propose that rather than stabilization, a thin and dense earth plaster can be use that protects earthen walls against erosion making the material more sustainable.

The mechanical property values of unfired bricks (CEBs) found in the literature are difficult to compare because of the use of different earths, sometimes of unknown composition, and variable or no stabilisation. Furthermore, the blocks are fabricated with different water contents, densities and additions. This is reflected in the great disparity of the strength values. However, in general, the values in the literature agree with the results of this investigation, where the compressive strength ranges from 2.2 to 5 N/mm² and the flexural strength varies between 0.48 and 1.53 N/mm². For example, Oti et al. [7], using Lower Oxford Clay (23% illite, 10% calcite), two different types of lime, Ground Granulated Blastfurnace Slag (GGBS) and Portland cement (PC) reached approximately 3.5 N/mm². González-López et al. [52] testing CEBs manufactured with silica and clay, and varying compaction, reached strengths of c. 2 N/mm² and maximum strengths over 11 N/mm² (for blocks stabilised with 15% PC). The results by Millogo et al. [65], who tested lime-stabilised (up to 12%), kaolinite-clay, reached 2.3-3.4 N/mm², and Toure et al. [70] who investigated a laterite soil stabilised with PC, reached 1.3–3.3 N/mm². These are comparable, although slightly lower than those in this paper. Similarly, Mansour et al. [83] also reached lower values, where the CEBs of high bulk density (equal to this study = 2200 kg m⁻³) reached up to 4 N/mm². In contrast, Sitton et al. [84] reached greater values (7-15 N/mm²) using a soil of unknown composition stabilised with 5-9% PC. Likewise, Teixeira et al. [85] also reached high strength values (7.8–11.0 N/mm²), likely triggered by a high compaction, the stabilisation and the high sand/pebble content in the clay (15.9% pebble, 47.2% sand, 17.6% silt, and 19.4% clay).

4.2. An estimation of the strength of masonry built with the unfired bricks

Costigan and Pavia [86], based on regression analysis of experimental data, proposed the equations in Table 11 to estimate the compressive strength of lime masonry based on the strength of the masonry units and the mortar. Taking the strength of the unfired illitic clay brick (4.98 N/mm²), and a standard hydraulic lime mortar of 3.5 N/mm² strength (NHL3.5), the masonry would reach a strength of 2.45 N/mm² which closely meets the structural requirements of EN 1996-2 [87] and BS 5628-1 [88] standards for fired-brick masonry. These standards include structural requirements where, for a brick of 5 N/mm² compressive strength bound with any mortar strength class, the characteristic masonry compressive strength is set at 2.5 N/mm². Furthermore, bricks are often nonstructural materials, and the loads are carried by frames.

4.3. Properties of the unfired brick vs the fired product

When comparing the properties of the fired and the unfired bricks (Table 12), it can be seen that the thermal conductivities are comparable. However, the unfired brick is more permeable to vapour which can improve indoor environment. Few standard requirements exist for bricks in construction, unless they are required for specific purposes where harsh conditions apply. It is evident that fired-clay bricks are mechanically stronger and more resistant to moisture, frost and salt damage than unfired clay bricks; and that these would rate low in existing durability standards because they are not suited for earthen materials. However, this doesn't imply low durability but, on the contrary, historic legacy demonstrates the long-lasting nature of earth construction.

In addition, in certain non-loadbearing applications such as internal partitions or rendered outer leafs of cavity walls or external walls, the bricks do not require structural strength and can be protected from weathering. Therefore, for certain applications, bricks do not require a set of specific requirements concerning durability but an adequate protection and maintenance to minimise long-term damage. As explained by Van Damme and Houben [18] as long as the earth properties are suited to the applied construction technology, and an impermeable plinth and foundation as well as a projecting overhang are in place, unstabilized earth structures are lasting, even in wet climate. Outdoor exposure of stabilized and raw earth walls conditions ensued a good durability [89].

According Van Damme and Houben [18] durability studies have concluded that unstabilized rammed earth walls can last for over 60 years without maintenance and without significant loss of strength safety factor, while still keeping their aesthetic value. However, the authors state that the 60-year value is too cautious and unrealistic, because it is based on the assumption of a linear, initially anomalously high, erosion rate. The non-linear erosion rate of earth materials has also been emphasized by other authors [92].

4.4. Potential impacts of the production of unfired illitic-clay units

As aforementioned, the illitic clay at Kingscourt brick is extruded, wire-cut and fired in tunnel kilns at 1030 °C. The producers currently spend €650k per annum on natural gas to dry and fire 14,000,000 bricks and emit 5000 tonnes of CO_2 to meet the

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Equations proposed for the estimation of the compressive strength of lime masonry based on the regression analysis of experimental data in Costigan and Pavia [86].

Mortar	Regression model for the estimation of masonry compressive strength
All limes (NHLs & CL90s)	$f_m^{'} = 0.56 f_b^{0.53} f_j^{0.5}$
NHL only	$f_{m}^{'} = 0.69 f_{b}^{0.55} f_{i}^{0.37}$
CL90s only	$f_{m}^{'}=0.55f_{b}^{-0.5}f_{j}^{-0.5}$

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Table 12

Material	Density Kg/ m ³	Compressive strength N/ mm ²	Thermal Conductivity W/m. K	Water Absorption %	Water vapour resistance factor
Illitic clay brick –fired at 1030 °C	1970	35 to 60	0.63	≤10-12	6–12 * 5–16.3**
Illitic clay unfired	c.2200	2.3 to 5	0.65-1.20	16-20	5.28-6.98



Fig. 9. World-wide distribution of the dominant clay mineral groups in the topsoil and subsoil in Ito and Wagai [90]. Reproduced with the authors' permission. Legend – 4: Illite, 3: Vermiculite, 2: Smectite, 1: Kaolinite.

demands of their existing market in Ireland and Europe. As a result of their emissions, in 2019, they spent approximately 115,000 euros in carbon tax which will raise to approximately 175,000 euros in 2020, as the carbon tax rises from \pounds 23/tonne to \pounds 35/tonne (R. Glennon pers.com. 2018). The bulk of their market is the con-

struction of domestic, commercial or industrial buildings where the bricks are used in internal and external walls, floors, arches and cornices, partitions and retaining walls. As seen above, the unfired units meet abundant regulations and can be used in certain applications where the fired bricks are currently used. In the hypothetic scenario where half of the current brick production was unfired to be used for specific applications, the producers would economise 325,000 euros in kiln fuel and 87,500 euros in carbon tax annually which, in 10 years, would amount to well over 4 million euros.

With respect to environmental impact, Kingscourt would produce approximately 2,500 tonnes of CO_2 per year (as opposed to 5000 tonnes of CO_2) if half of the production was to switch to unfired units. Kingscourt is a relatively small producer, but the impact would be gigantic if the large producers in China, Spain, India, and France (main brick producers in the world) switch part of their production to unfired units. For example, India (3rd world's largest producer) emits between 78 and over 200 million CO_2 eq. tonnes per year [6].

As evidenced by the results above, the illitic clay can be used unfired. The volume of illitic clav brick produced annually in the world could not be found, as most of the published literature does not report the exact composition of the clays used for brickmaking. However, the extent of illitic sediments is significant on the planet. According to Ito and Wagai [93] the most abundant clay mineral in the topsoil and subsoil in the world is illite (Fig. 9). As a result, Illitic clays are used for brick making all over the world. Most of the topsoil and subsoil of the world's top brick producers China, Spain, India and France are in illite clay areas [93]. In addition, some of the 10 leading US brick producer states (Texas, Alabama, Oklahoma, North Carolina, Oregon, Ohio, New York, California, Arkansas and Indiana, which accounted for 68% of common clay and shale sales [3] are also located in illite clay areas, as well as a significant number of producers in Ireland, Italy, UK, Germany, Greece, central and western Europe and most of Russia. This indicates that using illitic clay unfired world-wide would significantly lower the deterioration of the environment.

5. Conclusion

The results suggest that illitic clay can be used, unfired, in construction, either stabilized with lime or alone. All the clay's geotechnical parameters are within the limits considered suitable for earth construction by building standards and previous authors. The PI vs PL values meet the requirements of the Spanish standards for the fabrication of compressed earth blocks; and the granulometry lies within the values recommended for materials for earth construction.

The compressive strengths of all the mixes (both stabilized and raw), ranging from 2.20 to 4.98 N/mm², fall within accepted/recommended limits for earth construction in existing codes and standards, and the flexural strengths (0.48–1.34 N/mm²) are within the recommended values in earth building guidelines (USA, Australia and New Zealand).

Masonry constructed with the unfired units, bound with a standard hydraulic lime mortar would reach a strength of 2.45 N/mm² (28 day) which meets the structural requirements of some of the European standards for fired-brick masonry. When compared with the fired brick, the strength and durability are lower however, the vapour permeability is superior and the thermal conductivity comparable. Therefore, the unfired, illitic-clay units can be used in certain applications where fired ones are currently used, provided they are limited to suitable applications where the strengths reported above are contemplated and the material is sheltered against the effect of water.

The stabilization of illitic clay improves durability in liquid water but lowered strength (both compressive and flexural) and vapour permeability. However, all the water vapor resistance factors are within the 5–10 range recommended by the German standard on earth bricks. Lime stabilization increased the specific heat

capacity of the clay and tends to increase the thermal conductivity, thus enhancing the thermal mass but lowering the insulating capacity of the material.

The strength reduction caused by lime stabilization is attributed to a combination of factors including: the adsorption of the Ca^{2+} by the clay minerals (lime fixation) which prevents free Ca^{2+} from taking part in the pozzolanic reaction which creates strength; the competition for water between the hygroscopic clay minerals and the lime, and the short curing.

If half of the current production of Kingscourt brick (14,000,000 bricks per year) was unfired, the producers would economise over 4 million euros in kiln fuel and carbon tax in the next 10 years, and would produce approximately 2500 tonnes of CO_2 per year (as opposed to the 5000 tonnes of CO_2 that they currently produce). Kingscourt is a relatively small producer, but the impact would be vast if the main brick producers in the world would switch part of their production to unfired units. Illitic clays are used for brick making all over the world, and much of the soil of the world's top brick producers China, Spain, India and France is in illitic clay areas. Using unfired illitic clay world-wide would significantly lower the deterioration of the environment. For example, the 200 million CO_2 eq. tonnes emitted in India every year to produce bricks could be halved.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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