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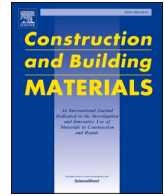
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Earth construction: Field variabilities and laboratory reproducibility

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ABSTRACT

Building construction is a major polluting sector. As a result, there is increasing global interest in the development of sustainable building materials with low environmental impact. Earth-based materials are among the materials of interest and building with earth-based materials has thus received a particular renewal of attention. Previous research has focused on the physical characteristics and durability of these materials. The aim of this study is to assess the variability of materials made *in-situ* and their reproducibility in the laboratory using an automatic normal Proctor machine with different compaction energies. Both cob and light earth were investigated. Cylindrical and prismatic specimens were produced on-site and in the laboratory: cob was made of silt, silty clay, sandy silt, and flax straw; and a separate layer of light earth was made of elastic silt and reed fibres. An experimental program was designed to evaluate the properties of the materials in terms of their water content, density, porosity, compressive strength, and thermal conductivity. The results revealed that the *in-situ* densities could be reproduced in the laboratory with compaction energies of 0.6 MJ/m³ and 0.2 MJ/m³ for cob and light earth, respectively. These compaction energies will allow the research to produce laboratory specimens that were representative of the materials implemented on-site. Regarding the compressive strength, the values obtained in the laboratory were higher than those of the *in-situ* specimens. Correction factors of 0.88 and 0.67 for cob and light earth. These values should be applied to calibrate the laboratory results in relation to *in-situ*. Concerning the thermal conductivity, the values obtained in the laboratory were similar for cob and higher for light earth. A correction factor of 0.87 should be applied to calibrate the laboratory results to those obtained *in-situ*.

1. Introduction

Most modern buildings are constructed with highly processed materials, which are responsible for a large proportion of greenhouse gas emissions. Broadly speaking, energy is consumed during the manufacturing, transport of these materials from production to building sites, construction phase, operating phase, and building demolition. The combination of these processes is responsible for more than 40% of global energy consumption, which severely depletes the world's natural resources [1]. Therefore, there is growing interest in the use of natural materials, particularly locally available earth building materials, which are minimally processed and generate low carbon emissions [2,3]. Earthen construction has recently received increased attention owing to its low environmental impact and recyclability [4-6]. Moreover, in 2022, France will adopt a new environmental regulation [7], which aims

at limiting the overall carbon footprint of new buildings and reduce energy consumption objectives.

Throughout the world, earth is used as a building material in a variety of forms, including rammed earth, light earth, adobe blocks, compressed earth blocks, and cob [8,9]. In the last three decades, many researchers have conducted detailed studies on the characteristic properties of rammed earth, adobe, and other compressed earth blocks [10-13]. This research has elucidated the thermal and mechanical behaviours of different earthen-based materials and techniques (see Tables 1 and 2).

As with other earth-building techniques, cob is currently attracting renewed interest and finding new applications in modern construction. This is largely due to its various advantages, particularly the comfort provided to occupants as well as its durability [14,15]. In France and the United Kingdom, cob remains one of the most common earth-building techniques. It is made with earth, water, and fibre (traditionally wheat

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Nomenclature			
ISM	In-situ mixes	M_t	total mass [kg]
LM	laboratory mixes	W	water content mass [%]
CEB	Compressed earth blocks	$W_{\text{Absorption}}$	absorption coefficient [%]
F_{content}	fibre content [%]	\varnothing	diameter of cylindrical specimens [mm]
I_p	plasticity index [%]	H	height of cylindrical specimens [mm]
W_L	liquid limit [%]	σ	maximum compressive strength [MPa]
MBV	methylene blue value [g/100 g]	$\sigma_{2\%}$	compressive strength at 2% [MPa]
M_{air}	mass of saturated sample in air [kg]	ε	Strain [%]
M_{oil}	mass of saturated sample in non-wetting oil [kg]	ρ_{Absolute}	absolute density [kg/m ³]
M_{dry}	mass of dry sample [kg]	ρ	bulk density [kg/m ³]
		Φ	Porosity [%]
		λ	thermal conductivity [W·m ⁻¹ ·K ⁻¹]

Table 1

Thermal conductivity of earthen-based materials in the literature.

Reference	Construction technique	Density [kg·m ⁻³]	λ [W·m ⁻¹ ·K ⁻¹]
Goodhew et al., 2021 [28]	Light earth	399	0.12
Colinart et al., 2020 [29]	Cob	1419	0.45
	Light earth	190–353	0.06–0.12
Vincelas et al., 2019 [30]	Lime-hemp	330–400	0.11
Azhary et al., 2018 [31]	Unfired clay bricks	1610–1890	0.35–0.46
Cagnon et al., 2014 [32]	Earth bricks	1900–2100	0.41–0.59
Röhlen et al., 2013 [33]	Compressed earth blocks	1600–2100	0.70–1.20
	Rammed earth	1600–2200	0.70–1.40
	Cob	1400–1700	0.60–0.80
	Earth coating	1000–1800	0.35–0.91
Minke, 2000 [34]	Cob	1200–2000	0.47–0.93

Table 2

Compressive strengths of cob reported in the literature.

Reference	σ [MPa]
Jiménez Rios and O'Dwyer, 2020 [35]	0.59–1.41
Quagliarini et al., 2018 [21]	1.12–1.35
Miccoli et al., 2017 [36]	1.59
Röhlen et al., 2013 [33]	0.6–1.3
Keefe, 2012 [24]	0.6–1.4
Saxton, 2006 [37]	1.65
Ziegert, 2006 [38]	0.7–1.2
Akinkurolere et al., 2006 [23]	0.6–2.2

straw [16,17]. In addition to its environmental advantages, cob also exhibits interesting behaviour when considering occupant health, indoor air quality, and social and convenience benefits [14,18]. Recently, several studies have been conducted on the characteristics of cob materials [14,19]. Other researchers have investigated the scale effect of cob properties. In addition, studies on cob walls under static and dynamic loads have been conducted to determine their performance [20–22]. A compressive strength of 0.6 MPa has been suggested to be sufficient to provide a margin of safety for cob dwellings of up to two stories in height [23]. In addition, the use of natural fibres provides tensile strength to cob structures [24]. In the literature, the average compressive strength of cob ranges from 0.60 to 1.65 MPa (see Table 2).

The investigation of engineering properties is important in modern design practices and code requirements. Therefore, cob needs standardisation in the laboratory to assess its field properties and *in-situ* monitoring to assess its material variability during construction. Several earth-building standards exist in New Zealand (NZS 4298) [25],

Australia (HB 195) [26], and India (IS 13827) [27], but they do not address cob construction.

2. Focus of this study

A prototype earthen-based building with a total area of 20 m² (indoor area of 13 m²) is under construction in Normandy (France). This prototype uses double-layer earth-fibre walls (Fig. 1) to comply with the French thermal regulation (RT 2012). The ground plan of the building includes two different layers in the walls: the inner side is cob, and the outer side is light earth (Fig. 2). Two different thicknesses are considered: 70 cm in the north and east walls to ensure better thermal insulation, and 50 cm in the south and west walls that are more exposed to solar radiation (Fig. 2). Earth-fibre walls are built on a 70-cm-high composite sub-base made of light concrete and cellular concrete. This sub-base structure ensures a similar thermal resistance of the earth-fibre walls. The prototype earthen-based building is also intended to establish various construction details for using these types of materials [39].

The present study focuses on the performance monitoring of the cob and light earth used in the construction of the prototype building from lift 2 to lift 5. The physical, mechanical, and thermal properties of the *in-situ* mixes (ISM) and laboratory mixes (LM) are evaluated and analysed. The aim of this study is to reproduce two different earth-based materials in the laboratory. The first is a cob made of silty clay, sandy silt, and flax straw. The second is light earth made of elastic silt and reeds. For this purpose, an automatic Proctor machine with different compaction energies (0.2, 0.3, 0.5, 0.6, and 0.7 MJ/m³) is used.

3. Materials and methods

3.1. Materials

The materials used for this study comprise three natural soils and two vegetable fibres from Normandy (France). The soils were characterised based on their granulometry and clay activity. Particle size analysis was performed by mechanical wet sieving for particles above 80 μm , according to standard XP P94-041 [40], and by laser granulometry for particles under 80 μm using an LS 13 320 from Beckman Coulter, following standard ISO 13320 [41]. The clay activity was evaluated according to the methylene blue value test (NF P94-068) [42] and Atterberg's limit (NF EN ISO 17892-12) [43]. The soil particle size distribution is shown in Fig. 3. These characteristics allow the classification of soils using the LCPC-USCS classification [44]: Soil 1 is a sandy silt, Soil 2 is a silty clay, and Soil 3 is an elastic silt (see Table 3).

In addition, two types of vegetable fibres were used: flax straw and reed. The fibres absolute density was determined using a helium pycnometer (Accupyc II 1340) [45]. The water absorption capacity was determined according to the RILEM protocol [46]. The fibre properties are listed in Table 4. After 24 h, the flax straw and reed showed water absorption capacities of 431% and 203%, respectively (see Table 4). The



Fig. 1. View of the earthen-based building under construction in Normandy (France).

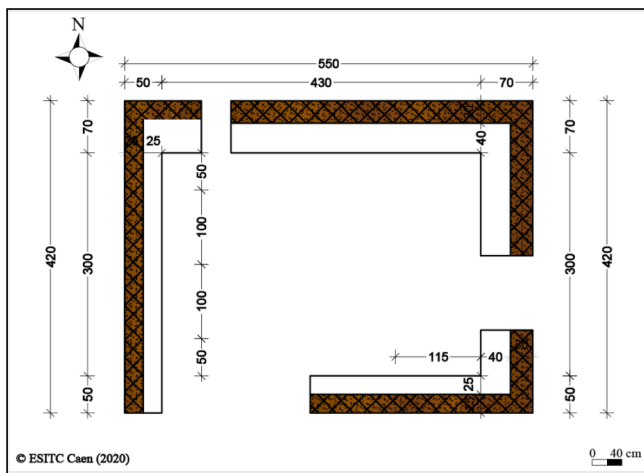


Fig. 2. Ground plan of the earthen-based building.

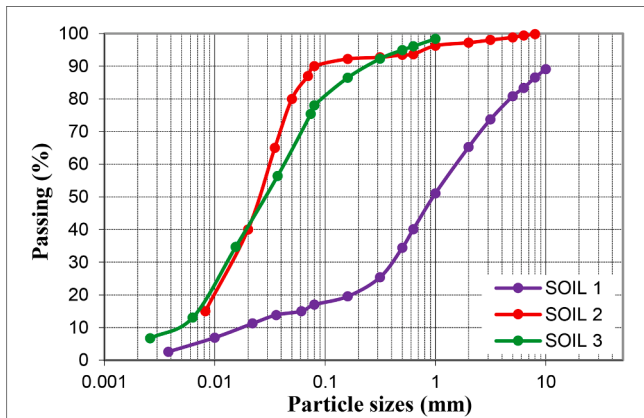


Fig. 3. Soil particle sizes distributions.

water absorption capacity of fibres influences the mix properties in the fresh state (absorption of available water) as well as in the long term (hygrometric balance).

3.2. In-situ mixes and casting details

To construct the prototype building, two different formulations of earth-fibre mixes are selected: the first is a cob made of silty clay, sandy

Table 3
Soil properties.

	Soil 1	Soil 2	Soil 3
Nature of soil (USCS classification)	Sandy silt	Silty clay	Elastic silt
W _i [%]	22.8	28.5	57.4
I _p [%]	2.3	4.2	14.9
MBV [g/100 g]	1.35	2.31	6.37
Particles < 80 μm [%]	17	90	78

Table 4
Vegetal fibre properties.

Fibre	Flax straw	Reed
ρ _{Absolute} [kg/m ³]	1266 ± 55	1305 ± 11
W _{Absorption at 24 h} [%]	431 ± 34	203 ± 9

silt, and flax straw. The soil 2 is very silty with a high plasticity index. In order to have a better mechanical behaviour, soil 1 was added as particles size corrector. The second is light earth made of elastic silt and reeds. The theoretical cob ISM is composed of 32.5% Soil 1, 65% Soil 2, and 2.5% flax straw (see Table 5). The theoretical ISM formulation for light earth is 66.7% Soil 3 and 33.3% reed (see Table 5). The different components and preparation tools are shown in Fig. 4.

In-situ cylindrical and prismatic specimens were prepared using a manual compaction rod. In total, 12 cylindrical specimens (Ø110 mm × H220 mm) were used to determine the bulk density, compressive strength, and fibre content. Twelve prismatic specimens (300 mm × 300 mm × 70 mm) were used to determine the bulk density and thermal conductivity. In addition, six large specimens (600 mm × 250 mm × 300 mm) were obtained by trampling and used to determine the bulk density, as shown in Fig. 4.

3.3. Laboratory mixes and casting details

To achieve the objectives of the present study, the properties of the LM should reproduce those of the ISM. To accomplish this, the LM should contain similar fibre and initial water contents as the ISM. Thus,

Table 5
ISM mix formulations of cob and light earth.

Cob			Light earth	
Soil 1 (Wt. %)	Soil 2 (Wt. %)	Flax straw fibre (Wt.%)	Soil 3 (Wt. %)	Reed (Wt. %)
32.5%	65%	2.5%	66.7%	33.3%



Fig. 4. In-situ preparation of cob and light earth.

the actual initial water content and fibre content were determined for the ISM specimens (see Section 4.1). Because of the flax straw high-water absorption, when preparing cob, a rest period is necessary to ensure a good water content homogeneity and cohesion in the mix.

Three cylindrical specimens of cob and light earth were prepared with different compaction energies using an automatic Proctor apparatus according to NF P94-093 [47]. The energy values used to compact the specimens were 0.2, 0.3, 0.5, 0.6, and 0.7 MJ/m³, as shown in Fig. 5. When performing compaction on both mixes, Proctor type A moulds were replaced with Ø110 mm × H220 mm cylindrical moulds.

3.4. Methods

3.4.1. Water content and bulk density

During construction (from lift 2 to lift 5), samples of cob and light

earth were taken from the site, and their real water content was measured according to standard NF ISO 11465 [48].

Cylindrical and prismatic specimens obtained from the building site and those prepared in the laboratory were kept at 20 ± 2 °C and a relative humidity of 50 ± 5 % for one day and then dried at 40 ± 2 °C [49] until reaching equilibrium (three consecutive weights at 24 h intervals within a standard deviation of 1%). The bulk density was determined following standard NF X31-501 [50].

3.4.2. Mechanical behaviour

The uniaxial compressive strength (UCS) was measured for cylindrical specimens (Ø110 mm × H220 mm) [51]. The tests were carried out using an IGM press machine with a capacity of 250 kN. The press was force-controlled with an imposed loading rate of 0.4 kN/s. The loading rate is chosen according to NF EN 13286-41 [52]. Indeed, the cracking in

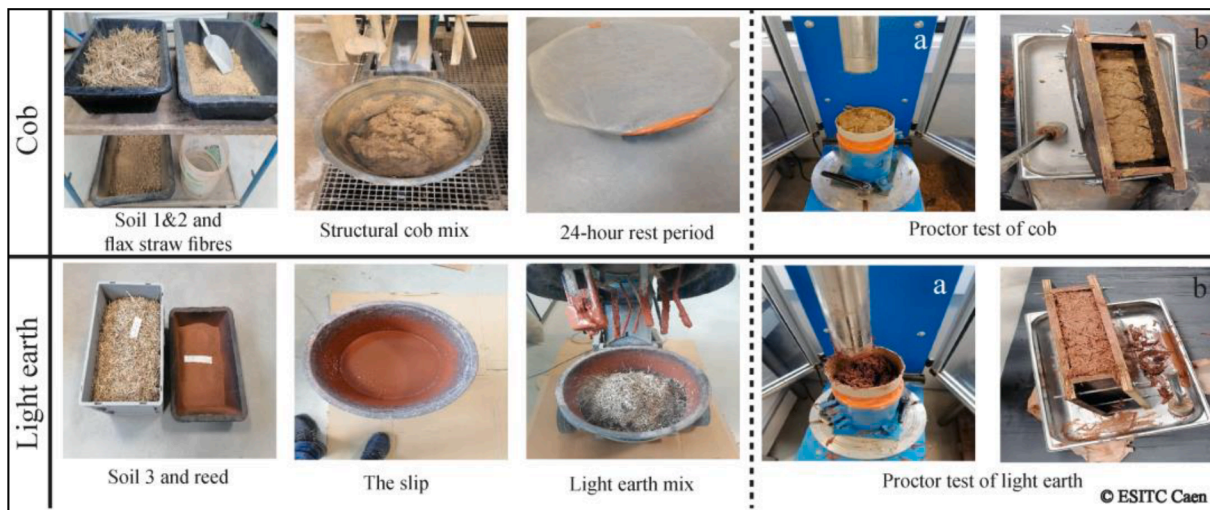


Fig. 5. Preparation of cob and light earth specimens in the laboratory: (a) cylindrical specimens for compressive strength tests and (b) prismatic specimens for thermal conductivity tests.

this standard need to be between 30 and 60 s. The value of 0.4 kN/s seems a good choice to face a mechanical strength between 1 and 2.5 MPa. The mechanical performance of the cob and light earth was determined for 2% strain [53,54]. This choice was made to provide representative of the building behaviour on site.

3.4.3. Thermal behaviour

The thermal conductivities of prismatic (300 mm × 300 mm × 70 mm) cob and light earth specimens were determined using a Netzsch HFM436 Lambda heat flux meter (HFM) according to ISO 8301:1991 [55]. This technique consists in applying a temperature gradient between two plates at a mean temperature of 20 °C (the cold plate is at 10 °C and hot plate is at 30 °C) to measure the heat flux through the studied material. When the equilibrium state is reached and the heat flux is constant, the thermal conductivity of the material is calculated using Fourier's law [56].

3.4.4. Accessible porosity

In this study, the accessible porosity was measured by immersing small samples in non-wetting oil (dearomatized oil) according to standard NF ISO 5017 [57]. The samples were dried in an oven at 40 ± 5 °C, placed in a non-wetting oil-filled beaker, and saturated under vacuum in a desiccator for at least 24 h. This allowed the non-wetting oil to replace air in the open pores without interacting with the sample volume. Then, the specimens were weighed under air and under non-wetting oil (See

Fig. 6), and the accessible porosity was determined according to the following equation [57]:

$$\Phi = \frac{M_{air} - M_{dry}}{M_{air} - M_{oil}} \times 100 \quad (1)$$

where

- Φ is the porosity [%]
- M_{air} is the mass of the saturated sample in air [kg]
- M_{oil} is the mass of the saturated sample in non-wetting oil [kg]
- M_{dry} is the mass of dry sample [kg]

4. Results and discussion

First, the *in-situ* monitoring results are presented, and these are then used to determine the appropriate laboratory formulations. Then, the results will be presented from an automatic Proctor machine using different compaction energies to reproduce a series of *in-situ* densities in the laboratory. Finally, the differences between the LM and ISM properties are investigated to predict future mixes.

4.1. In-situ monitoring

4.1.1. Real fibre content

To monitor the variation in fibre content in the cob during the building phase from lift 2 to lift 5, an experimental protocol was implemented. First, the ISM cylindrical specimens (Ø110 mm × H220

mm) were dissolved in water. Then, the dissolved specimens were sieved at 6.3, 5, 3.15, 2.5, 0.9, and 0.5 mm, as shown in Fig. 7. The results reported in Table 6 indicate that the flax straw content varied between 2.1% and 2.3%.

For the light earth material, *in-situ* mixing was performed in three stages: Soil 3 was first mixed with water to obtain slip, then reed was added to the slip; finally, all of the materials were mixed. The reed fibre and soil contents were determined according to the buckets used *in-situ*, as shown in Table 7. The soil and fibre contents in the final specimens are given in Table 8.

According to the results reported in Tables 6 and 7, laboratory mixes were formulated with a flax straw content of 2.2 % for cob and a reed content of 36 % for light earth. The synthesis of the soil and fibre contents in both the cob and light earth are presented in Table 8.

4.1.2. Real initial water content

During the four lifts, the water content in the ISM was between 18% and 21% for cob and between 79% and 83% for light earth, as shown in Fig. 8. Compared to an average value, the results show that the variation in water content could reach approximately 7.7% for cob and 2.5% for light earth during the building phase.

Water content variation can occur as a result of sun exposure (especially for light earth) and approximations in the amount of added water (especially for cob). The water content variation between specimens can occur because large-size moulds are used after casting the cylindrical/prismatic specimens (Fig. 8b).

According to the results in Fig. 8, the laboratory specimens are formulated with an initial water content of 20% for cob and 80% for light earth. The initial water content percentage is related to the dry mass of the soil–fibre mixtures.

4.2. Bulk density

The *in-situ* specimen bulk densities are presented in Fig. 9 for different lifts and specimen shapes. The results indicate that the average bulk density of cob is similar regardless of the specimen shape: between 1740 kg/m³ for prismatic specimens and 1783 kg/m³ for large-size moulds. However, it is slightly different for light earth, with a mean of 487 kg/m³ for cylindrical/prismatic specimens and 443 kg/m³ for large-size moulds (Fig. 9a). This variation in bulk density may be due to the volume of the specimen, which is greater when using large-size moulds. It is thought that the fibre elasticity may lead to an immediate swelling after compaction that is proportional to the specimen size.

Analysis of the average bulk density for varying lifts with cylindrical/prismatic specimens shows that the bulk density is between 1691 and 1789 kg/m³ for cob and between 420 and 565 kg/m³ for light earth (Fig. 9b). When considering the different lifts, a bulk density variation of 3% for cob and 15% for light earth can be observed, as shown in Fig. 9b. The large bulk density variation in light earth may be due to the water content, which varies from 79% to 83%.

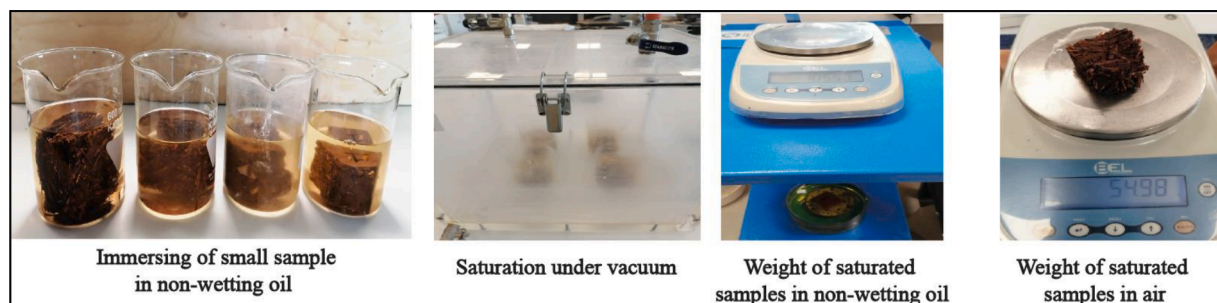


Fig. 6. Porosity measurement protocol.

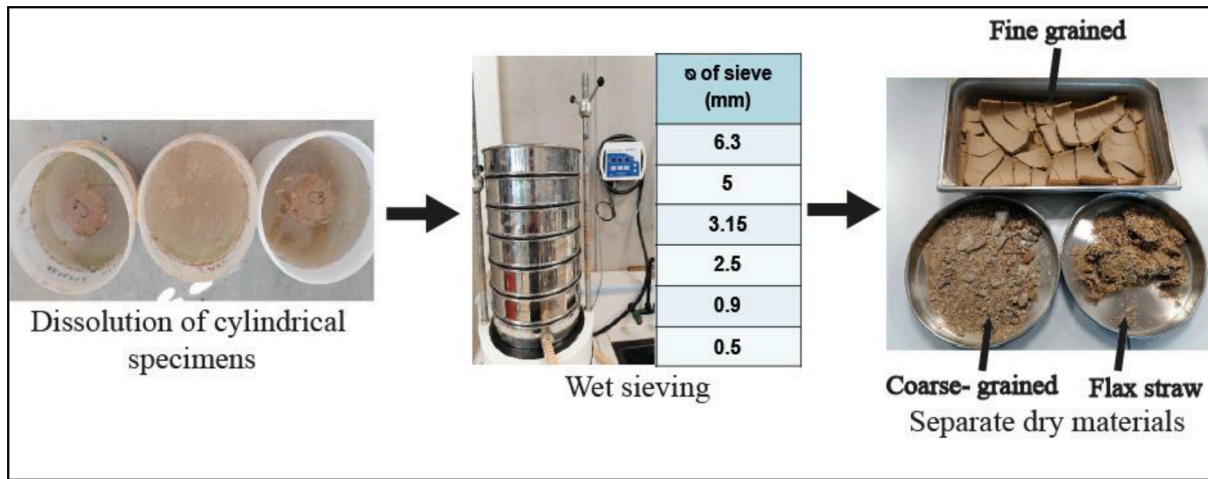


Fig. 7. Fibre and soil separation protocol.

Table 6
In-situ cob fibre contents.

Lifts	F _{content} [%]
Lift 2	2.3 ± 0.2
Lift 3	2.1 ± 0.2
Lift 4	2.3 ± 0.1
Lift 5	2.2 ± 0.2

Table 7
Soil and reed contents in light earth.

Material	Number of buckets	Mass of bucket [kg]	Total mass [kg]	Content [%]
Soil 3	4.5	13.6	61.2	64
Reed	4.5	7.5	33.8	36

Table 8
Soil and fibre contents in cob and light earth specimens prepared in the laboratory.

Cob			Light earth	
Soil 1 (Wt. %)	Soil 2 (Wt. %)	Flax straw (Wt. %)	Soil 3 (Wt. %)	Reed (Wt. %)
32.6%	65.2%	2.2%	64%	36%

4.3. Mechanical behaviour

The compressive strengths of the specimens from different lifts are shown in Fig. 10. In the case of cob, the compressive strength at 2% strain varies between 0.96 and 1.35 MPa. These values are in accordance with those in the literature (see Table 2). The variation in compressive strength between the different prototype lifts is 16%. For light earth, the measured values are between 0.03 and 0.12 MPa. As these values are low, no conclusions can be drawn.

4.3.1. Thermal behaviour

In the literature, several hygrothermal studies have been conducted on rammed earth and other forms of earthen construction techniques [14,37,65]. Fewer investigations have been undertaken on the thermal properties of cob and light earth (see Table 1). The results are presented in Figs. 11 and 12. Except for lift 4 with a value of 0.836 W·m⁻¹·K⁻¹, the cob thermal conductivity is quite similar in the other lifts (between 0.610 and 0.650 W·m⁻¹·K⁻¹). For light earth, the thermal conductivity varies from 0.132 to 0.146 W·m⁻¹·K⁻¹ (Fig. 12). When considering the four lifts, the cob and light earth thermal conductivities present a variation of 5%.

4.4. Laboratory reproduction

The laboratory formulations reported in Table 8 were used to study the ability of the laboratory results to reproduce those obtained for the in-situ specimens. To achieve this, an automatic Proctor machine was used to compact the specimens by applying different numbers of blows corresponding to different compaction energies (See Table 9).

The Proctor type A mould was replaced with a cylindrical mould

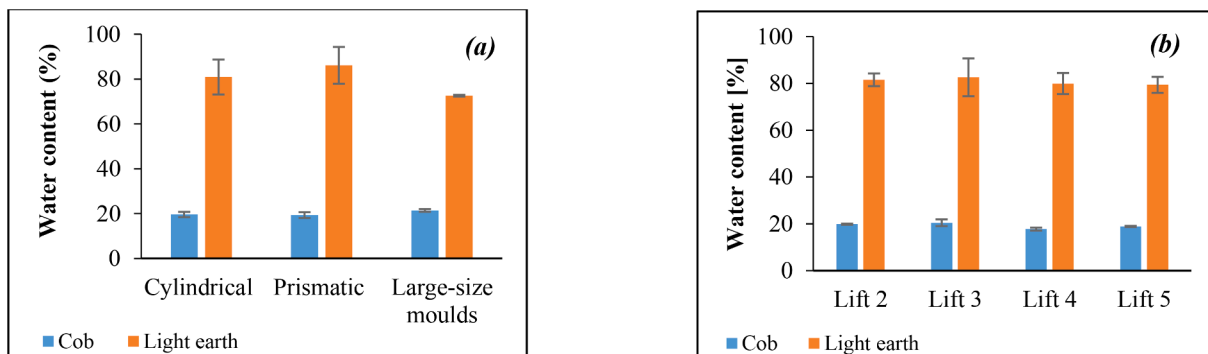


Fig. 8. ISM water content for different (a) specimen shapes and (b) lifts.

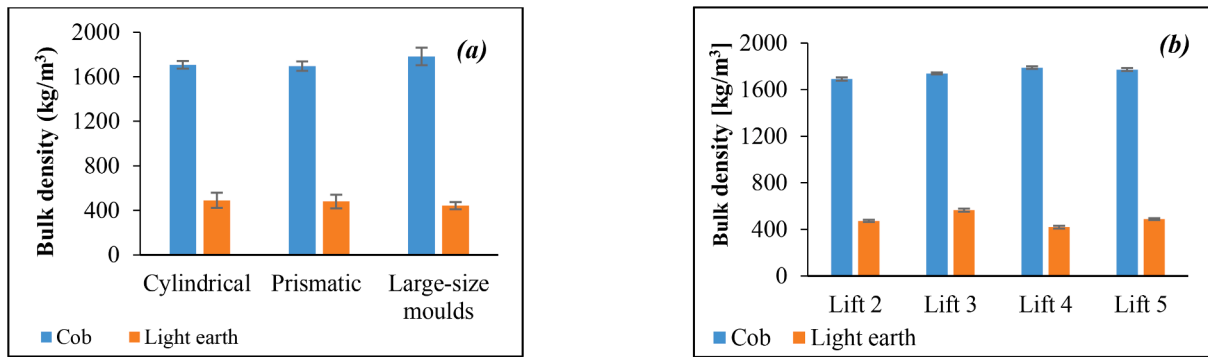


Fig. 9. ISM bulk density for different (a) specimen shapes and (b) lifts.

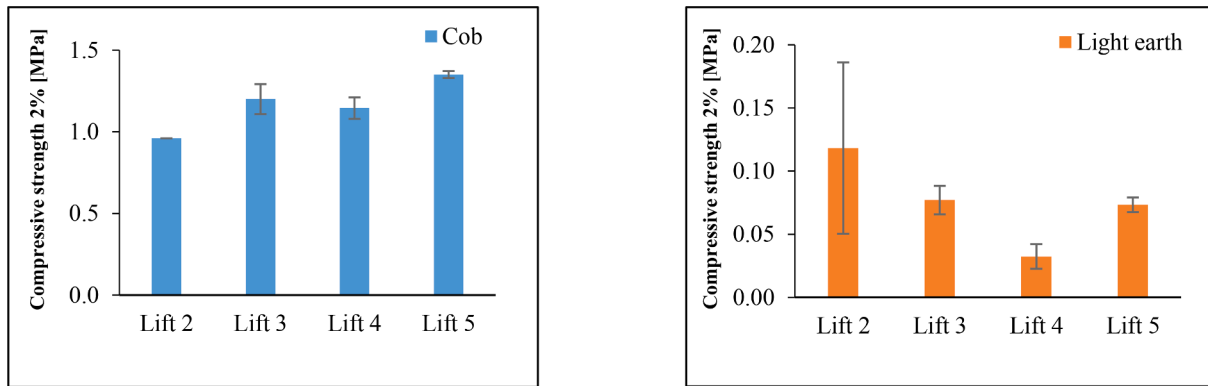


Fig. 10. ISM compressive strengths at 2 % strain.

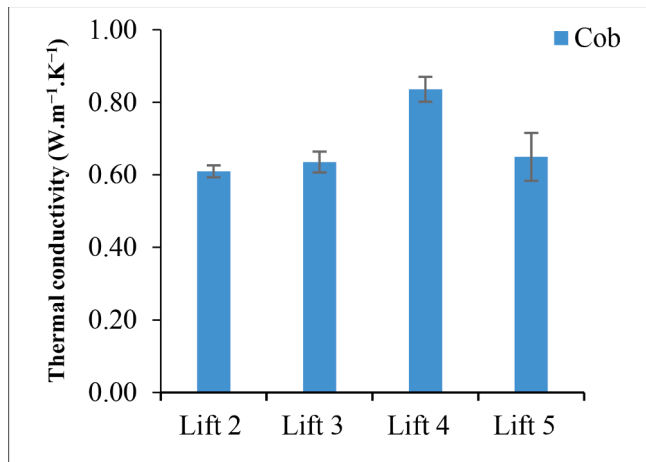


Fig. 11. Thermal conductivity of the *in-situ* cob.

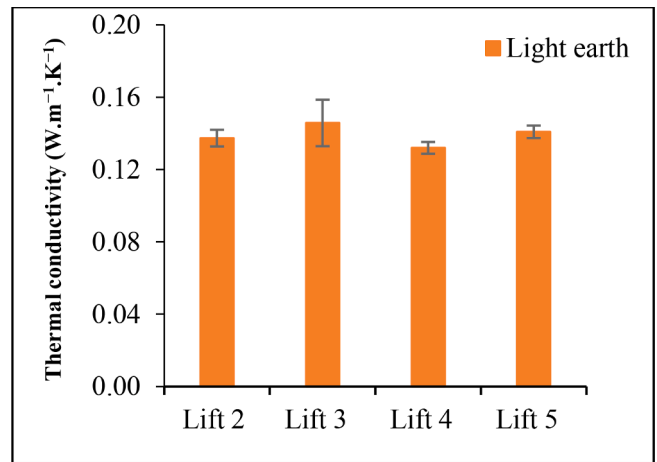


Fig. 12. Thermal conductivity of the *in-situ* light earth.

similar to that used for the compressive test specimens (Ø110 mm × H220 mm). The bulk density was used to determine the compaction energy that reproduced the ISM. The obtained results are shown in Fig. 13. It can be observed that the reproduction of the cob *in-situ* bulk density in the laboratory required a compaction energy of 0.6 MJ/m³. For light earth, a compaction energy of 0.2 MJ/m³ was needed.

Although the bulk density can be reproduced, the compaction differs between *in-situ* and laboratory specimens. Indeed, using an automatic Proctor machine will produce homogeneous compaction, which is not the case *in-situ*. Therefore, the accessible porosities of ISM and LM were measured (see Table 10).

Table 9
Laboratory compaction energies.

Number of blows	8	14	20	26	32
Compaction energy (MJ/m ³)	0.2	0.3	0.5	0.6	0.7

The results show that the cob specimens exhibited similar porosity, whereas there was a significant difference between the ISM and LM of light earth. This increase in accessible porosity can lead to lower compressive strength and higher thermal conductivity.

To investigate the impact of specimen homogeneity, the mechanical and thermal properties of the ISM and LM were determined and are

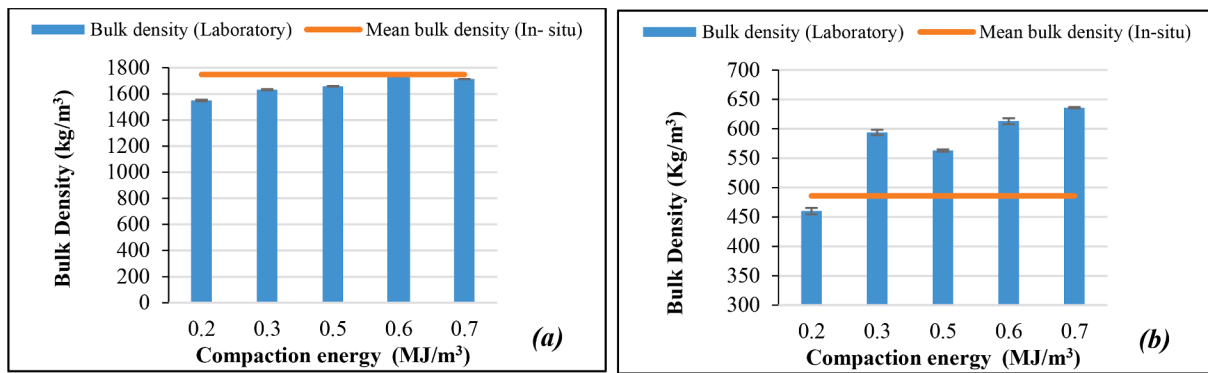


Fig. 13. ISM and LM density analysis: (a) cob and (b) light earth.

Table 10
Accessible porosity measurements for ISM and LM.

Material	Porosity “Φ” [%]	
Cob	ISM	30 ± 1
	LM	29 ± 3
Light earth	ISM	64 ± 2
	LM	53 ± 3

presented in Table 11. The homogeneous compaction led to an overestimation of the mechanical resistance and thermal conductivity for both the cob and light earth. Therefore, correction factors are proposed for each mix type and property (see Table 11).

5. Conclusion

This study focused on an investigation of the physical, mechanical, and thermal properties of cob and light earth.

Cob was made of silty clay, sandy silt, and flax straw, and light earth was made of elastic silt and reed. These two materials were used in the construction of a prototype earthen building in Normandy (France).

First, *in-situ* mixes (ISM) were analysed to evaluate the variability in properties between four lifts of the prototype earthen building. Regarding cob, the fibre content varied from 2.1% to 2.3%, and the water content varied from 18% to 21%; the bulk density values were between 1691 and 1789 kg/m³. The compressive strength at 2% strain varied between 0.96 and 1.35 MPa. This represents a 16% variation in the compressive strength of the cob during the prototype construction. Except for lift 4, the thermal conductivity ranged from 0.610 to 0.650 W·m⁻¹·K⁻¹. The water content of light earth varied from 79% to 83%, and the bulk density values were between 420 and 565 kg/m³. Regarding the thermal conductivity of the light earth, it varied from 0.132 to 0.146 W·m⁻¹·K⁻¹, which represents a 5% variation in the thermal conductivity of light earth during the prototype construction.

Second, an automatic Proctor machine was used with various compaction energies to prepare specimens in the laboratory. The laboratory mix (LM) formulations were based on the average percentages of materials used in the ISM. Bulk densities were used to determine the

Table 11
Laboratory mix correction factors.

Materials	σ _{2%} (MPa)	Mechanical correction factor	Mean λ [W·m ⁻¹ ·K ⁻¹]	Thermal correction factor	
Cob	ISM	1.35 ± 0.02	0.88	0.683 ± 0.054	0.96
	LM	1.54 ± 0.02			
Light earth	ISM	0.08 ± 0.01	0.67	0.139 ± 0.009	0.87
	LM	0.12 ± 0.03			

compaction energy, allowing similar materials to be obtained in the laboratory and on-site. A compaction energy of 0.6 MJ/m³ was found to be suitable for cob and 0.2 MJ/m³ was suitable for light earth.

Then, the mechanical and thermal properties of the LM were determined. The values obtained for the LM differed from those of the ISM. Indeed, the compressive strength and thermal conductivity of the LM were higher than those of the ISM. This is believed to be because Proctor compaction makes the material more homogeneous. Therefore, correction factors need to be applied to laboratory values to predict the *in-situ* values.

To continue this work, other mixes need to be studied to confirm the compaction energies. Concerning the ISM, the results obtained confirm that there is a need to perform on-site tests to ensure material performance and to assess material homogeneity.

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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