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Development of porous fired clay bricks with bio-based additives: Study of the environmental impacts by Life Cycle Assessment (LCA)

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HIGHLIGHTS

- Vegetable and chemical additives have been incorporated to the clay matrix.
- Physical, mechanical and thermal properties of the bricks have been characterized.
- Lighter and more insulating bricks were developed (best results for 1 wt.% WS).
- A Life Cycle Assessment of the developed materials has also been conducted.
- The incorporation of additives reduced the environmental impacts of the bricks.

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ABSTRACT

The incorporation of bio based pore forming agents, from either agricultural (wheat straw WS and olive stone flour OSF) or chemical (glycerol carbonate GC and dimethyl carbonate DMC) origins, into clay formulations, has been investigated. Fire clay porous tablets have been manufactured at laboratory scale and characterized through physical, mechanical and thermal properties. An increase of 7.2% of the porosity correlated to a decrease of 7.0% of the thermal conductivity was measured for the best samples.

The environmental impacts of these formulations have also been examined through a Life Cycle Assessment (LCA) using the ReCiPe v1.10 method. It was noticed that the incorporation of pore forming agents led a decrease of about 15–20% of all the studied impact categories.

The advantage of the use of bio based additives in clay bricks was then confirmed from both performance (lighter material with a better thermal insulation) and environmental points of view.

1. Introduction

Clay bricks have been widely used for thousands of years as it is an economic product using abundant and cheap raw materials (clay, sand and water) and through a simple manufacturing process [1]. However, since the arrival in the 1980s of the concrete block, the brick market began to decline. Brick producers were then facing technological barriers due to their limited insulating properties (0.12 W/m.K for the brick against 0.08 W/m.K for the concrete), and weight limiting their use to low rise buildings.

Nowadays, in a context of sustainable development and with the thermal regulations, it is necessary to develop new construction

materials with high thermal and mechanical performances. The incorporation of wastes from various origins has been evaluated to improve these properties. Indeed, the thermal decomposition during the process (drying and firing steps) of the pore forming agents leads to an increase of the porosity of the material and thus an increase of the insulation capacity [2–4].

But with today's environmental concerns (the global warming, the increase of ozone layer or even the accumulation of waste for example), the actors of building industry are increasingly forced to consider new ways to reduce their energy consumption. Indeed, in Europe, the building sector is responsible for 40–45% of primary energy consumption, contributing to significant emissions of greenhouse gases [5]. It thus becomes necessary to assess the environmental impacts of building materials by the Life Cycle Assessment (LCA). Many scientific studies dealing with the LCA of these products are designed to compare different materials

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together to highlight those with the least impact on the environment, whose majority is listed in the bibliographic review of Cabeza et al. [6].

Studies based on fired clay bricks were focused on determining the stages of the life cycle presenting the most important impacts on the environment [7–10]. Regarding this proper work about the development of new building blocks with biobased pore forming agent, very few data are available in the literature from the Life Cycle Assessment point of view. Two competitors of our industrial partner TERREAL have been identified as using biobased materials in their clay formulations (i.e. sawdust). Both have performed LCA through their Environmental and Health Declaration Sheets [11,12].

In this work, we studied the incorporation of additives from agricultural resources (wheat straw and olive stone flour) and bio based chemicals (glycerol carbonate and dimethyl carbonate). The two first ones were selected due to their low cost, availability and close location, and were already used as pore forming agents in our previous works [13]. The last two were chosen because their carbonate function easily degrades under thermal effect by liberating carbon dioxide and thus possibly creating pores [14].

The incorporation of these four pore forming agents in the clay matrix has been investigated including their effect on the various properties of clay materials. In addition, an environmental analysis, thanks to a Life Cycle Assessment (LCA), has been conducted to confirm the interest of adding biomass in these new microporous materials.

2. Methods

2.1. Development of fired clay bricks with biobased pore forming agents

2.1.1. Sample preparation

The characterization of clay and agricultural by products, from either chemical or physical point of view, was presented in a previous publication [13].

The clay, provided by TERREAL (Castelnaudary, France), is first ground to obtain a powder with particles of about 3 mm. Two different types of additives were incorporated and mixed with clay in a rolling mill to enhance homogeneity: wheat straw and olive stone flour called vegetable additives, glycerol carbonate and dimethyl carbonate called biobased chemicals. Wheat straw, provided by ARTERRIS (Castelnaudary, France), was crushed and sieved to obtain a grinding inferior to 0.5 mm (<0.5 mm). The olive stone flour, provided by BARDON ETS (Le Muy, France), was industrially ground at about 50 μm . As determined previously [13], wheat straw and olive stone flour are mostly composed of fibers (more than 80%) but present different mineral contents (8.89% for wheat straw and 0.99% for olive stone flour). Moreover, the characterization of the affinity to water shows that the water sorption and the swelling *ratio* were more important for the straw.

The amount of additive incorporated was chosen according to previous works [15]: 1 wt.% for wheat straw, 2 wt.% for olive stone

flour and 2 wt.% for both glycerol carbonate and dimethyl carbonate. The required quantity of water was added to obtain the desired humidity and plasticity that are necessary to avoid defects onto the structure during the process. The samples were then molded by extrusion process in the form of tablets ($175 \times 79 \times 17 \text{ mm}^3$), dried up to 105 $^{\circ}\text{C}$ and finally fired following a rise of temperature of 11 h until the maximum temperature of 920 $^{\circ}\text{C}$, maintained for 1 h, according to the industrial recommendations.

Samples were prepared and designated depending on the pore forming agent used: WS (wheat straw), OSF (olive stone flour), GC (glycerol carbonate) and DMC (dimethyl carbonate). The samples without additive are designated as reference samples (Ref.).

The samples composition is presented in Table 1.

2.1.2. Characterization of the bricks

The physical (linear shrinkage, loss on ignition, density, water absorption and porosity), mechanical (bending strengths) and thermal properties of the obtained clay bricks were determined.

Linear shrinkage was determined by measuring the length of the sample before and after drying using a caliper according to the standard of American Society for Testing and Materials C120 [16]. Loss on ignition was determined by measuring the mass loss of the sample between the drying and firing steps.

The bulk porosity and saturated density were determined according to the test procedure recommended by Hornain [17] by means of water saturation under vacuum. Once the samples were completely saturated, they were weighted then dried until constant mass and reweighted.

The bending strength of the fired samples was determined by three point bending with a loading rate of 0.5 kN/s until failure. Bending stress (σ_f N/mm² or MPa) was obtained through the following equation:

$$\sigma_f = \frac{3 \times F_{\max} \times l}{2 \times b \times h^2}$$

with F_{\max} the maximum force (N), l the support span (mm), b the section depth (mm) et h the section width (mm).

Thermal conductivity was obtained through a heat flux meter method. This method followed the standards of American Society for Testing and Materials C518 [18], ISO 8301 [19] and NF EN 12667 [20]. The measurement area was $60 \times 40 \text{ mm}^2$ with a thickness of the sample greater than 10 mm. This apparatus produced a temperature gradient along the thickness of the sample and measured heat flux that gave through the software the thermal effusivity. Thermal conductivity (λ W/m.K) was deduced using the following formula:

$$\lambda = \frac{E^2}{\rho \times C_p}$$

with E the thermal effusivity of the sample (J/m².K.s^{1/2}), ρ the bulk density (kg/m³) and C_p the specific heat capacity (J/kg.K).

All the characterizations were performed on 6 samples; the coefficient of variation of all the obtained values, defined as the

Table 1
Sample compositions.

	Ref.	WS	OSF	GC	DMC
Wheat straw (wt.%)	–	1	–	–	–
Olive stone flour (wt.%)	–	–	2	–	–
Glycerol carbonate (wt.%)	–	–	–	2	–
Dimethyl carbonate (wt.%)	–	–	–	–	2
Water (wt.%)	13.8	15.9	15.3	15.8	16.2

ratio of the standard deviation to the mean, is verified to be prior to 5%, showing the accuracy of the presented data.

2.2. Life Cycle Assessment (LCA)

The Life Cycle Assessment was undertaken using the ISO 14040 [21] and ISO 14044 [22] standards, the first one defining the principles and the framework and the second describing the different stages of the analysis.

2.2.1. Goal and scope definition

The Life Cycle Assessment was conducted on the entire manufacturing process of clay tablets in order to analyze and compare the environmental impacts of different formulations and identify the process unit that presents the strongest environmental impacts in an eco design approach.

In order to build the inventory of production and thus to set the scope of the study, the functional unit is defined, according to the works of Pargana et al. [23] as the production of 1 m² of porous tablets with a fixed thermal resistance that will be detailed later.

In order to focus only on the impacts associated to the development of these new samples, a cradle to gate analysis centered on both raw material extraction and manufacturing has been performed.

The studied system takes into account the production of raw materials (clay, vegetable and chemical adjuvants), the production of the energy consumed and finally the production, in itself, of tablets.

To overcome potential constraints, the initial hypotheses are defined as follows:

- The necessary infrastructure is not taken into account, thereby excluding their manufacturing and their dismantling;
- The electricity used considers the production mix corresponding to the French energy production system;
- The cleaning of the different devices used in the process (mill or extruder for example, as presented in Section 2.1.1.) is neglected;
- The synthesis of the chemical additives (glycerol carbonate and dimethyl carbonate) is assumed to be complete (yield = 1);
- The mass allocation is selected for the impact of the production of olive oil with 76% for pulp and 24% for the stone [24];
- The process being carried out locally, the transportation of the clay from career to the experimental site is neglected. On the contrary, the one for vegetables additives, wheat straw and olive stone flour, is taken into account.

2.2.2. Life Cycle Inventory

2.2.2.1. System description. For the Life Cycle Inventory, all the inputs and outputs of the system were listed for the different stages of the life cycle. The flowchart, presenting the different steps of the process with the associated flows, is presented in Fig. 1.

Noteworthy, the inputs, also called foreground data, detailed in Fig. 1, have their own life cycle. Those environmental impacts (background data) are taken into account for the realization of the global Life Cycle Assessment of the product.

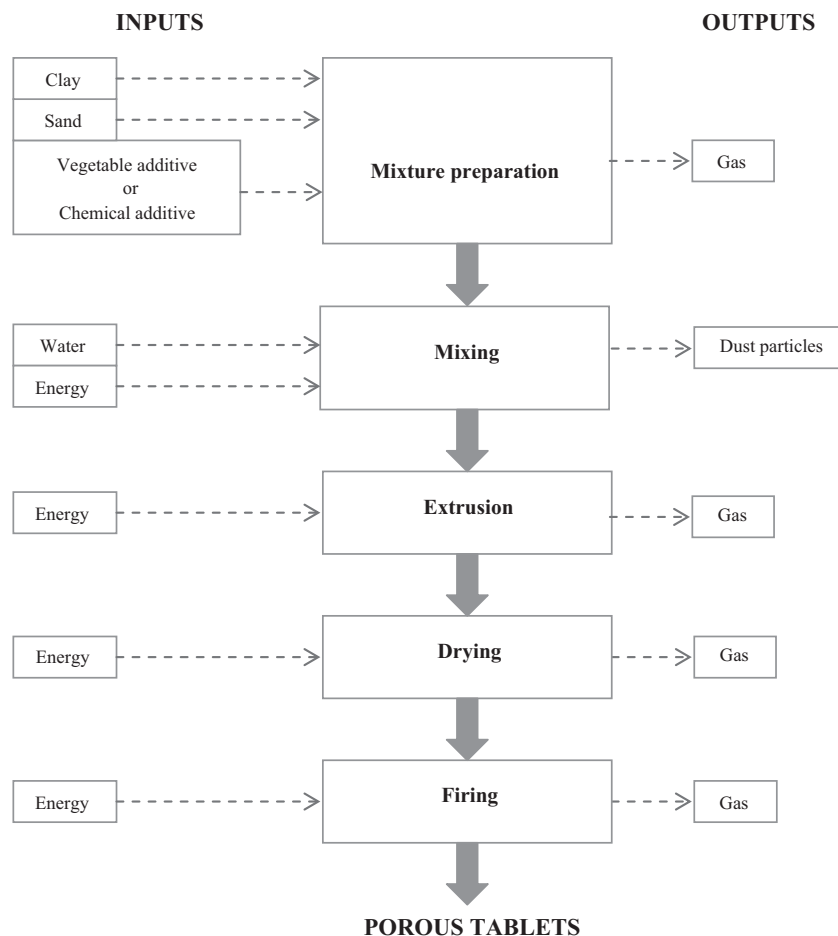


Fig. 1. Process flowchart of the production of porous tablets.

Table 2

Inventory data for the developed biobased bricks (the inputs followed by * are considered with their entire life cycle assessment).

	Ref.	WS	OSF	GC	DMC
Clay mixture (clay + sand)* (kg)	18.396	18.205	18.013	18.396	18.396
Straw* (kg)	-	0.125	-	-	-
Olive stone flour* (kg)	-	-	0.250	-	-
Glycerol carbonate* (kg)	-	-	-	0.240	-
Dimethyl carbonate* (kg)	-	-	-	-	0.240
Tap water (kg)	2.208	2.477	2.417	2.498	2.498
Electricity (kWh)	85.015	85.015	85.015	85.015	85.015

2.2.2.2. *Data collection.* The inventory data were obtained either directly from the experiments or by the use of data collected from the industrial partners or the literature (Table 2).

Data for the extraction of clay mixture, composed of clay and sand, were supplied by our industrial partner TERREAL (dating from June 2010). These data take into account the mineral extraction, the transportation from the clay quarry to the production site and all the energy consumptions associated to these processes.

For the vegetable additives, on the first hand, the cooperative ARTERRIS provided different fertilizer consumption in the cultivation of wheat (nitrogen, phosphorus and potassium), the amount of straw produced per hectare and the duration of the different stages of the process (harvest and bailing). On the other hand, for the olive stone flour, BARDON ETS provided the energy consumption associated with the grinding of the stones. As the olives directly arrived in the company premises, no data on the culture was available. A complete Life Cycle Inventory of olive cultivation, before processing, was collected from literature [25].

Due to confidentiality issues, all the process data provided by the industrials cannot be detailed in this publication either for the clay mixture or the vegetable pore forming agents.

As regards chemical additives, neither glycerol carbonate nor dimethyl carbonate were described in the database Ecoinvent

3.01; it was therefore necessary to model them under the software SimaPro 8.0. The glycerol carbonate was modelled according to the reaction presented by Claude et al. [26], while the dimethyl carbonate was modelled following the chemical pathway described by Murugan and Bajaj [27], as presented respectively in Figs. 2 and 3.

2.2.2.3. *Uncertainty analysis.* In this study, all data are not of equivalent accuracy and uncertainty exists for each of them. Therefore the quality of the data has been estimated precisely in order to guarantee the value used, and thus the interpretation of the results.

Quality indicators used are those developed by Weidema and Wesnaes [28]. For each data set, six parameters (U_1 to U_6) were qualitatively evaluated on a scale between 1 and 5 (1 being the better score and 5 the worse). The variance with a 95% confidence interval ($V_{95\%}$) is thus calculated according the following formula:

$$V_{95\%} = \exp\sqrt{\sum_i \ln(U_i)^2}$$

with U_1 the uncertainty factor related to the reliability parameter, U_2 the uncertainty factor related to the exhaustivity parameter, U_3 the uncertainty factor related to the temporal correlation parameter, U_4 the uncertainty factor related to the geographical

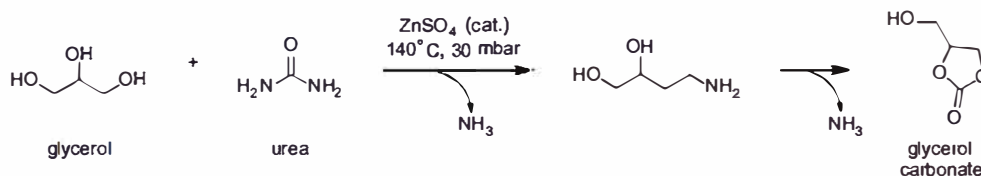


Fig. 2. Chemical formation of glycerol carbonate (adapted from Claude et al. [27]).

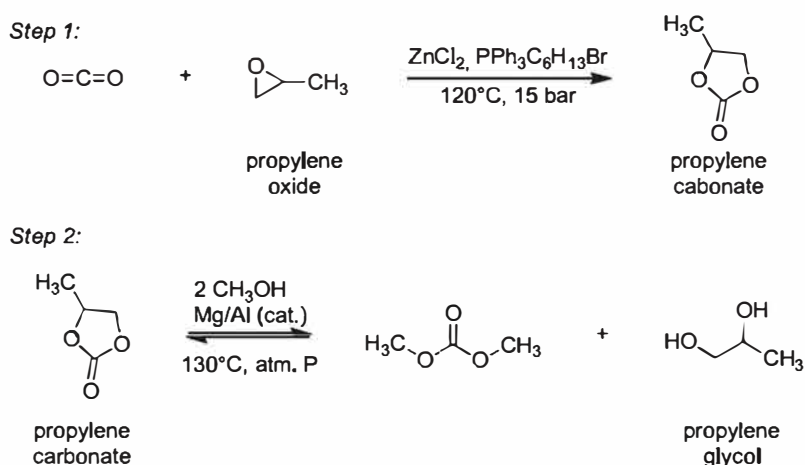


Fig. 3. Chemical formation of dimethyl carbonate (adapted from Murugan and Bajaj [28]).

Table 3
Midpoint impact categories used for the ReCiPe 1.10 calculation method.

Midpoint impact category	Description	Unit
Climate change	Emissions of greenhouse gases that cause an increase in temperature of the lower atmospheric layers (for example CO ₂ , CH ₄ , N ₂ O, CFC, CO...)	kg CO ₂ eq
Human health	Air emissions of substances that destroy the stratospheric ozone layer (for example CFC, HCFC, CCl ₄ ...)	kg CFC 11 eq
Ozone depletion		
Human toxicity	Emissions to soil, water and air of substances that harm human health (for example heavy metals, dioxins, VOC, NOx, SO ₂ , particulates...)	kg 1,4-DB eq
Photochemical oxidant formation	Air emissions of substances that cause the production of tropospheric ozone or smog (for example NOx, VOC, CH ₄ , CO...)	kg ethylene eq
Particulate matter formation	Air emissions of particulate matter less than 10 µm	kg PM10 eq
Ionising radiation	Ionising radiation or radioactive	kg ²³⁵ U eq
Change climate Ecosystems	Emissions of greenhouse gases that cause an increase in temperature of the lower atmospheric layers (for example CO ₂ , CH ₄ , N ₂ O, CFC, CO...)	kg CO ₂ eq
Terrestrial acidification	Air emissions of substances that cause acid rain (for example NOx, SO ₂ , NH ₃ , VOC, HCl...)	kg (SO ₂) ² eq
Freshwater eutrophication	Emissions to air and water of substances that cause an excess of nutrients in lakes, rivers and oceans (for example components containing N and P)	kg (PO ₄) ³ eq
Terrestrial ecotoxicity	Emissions to water and air of substances that harm living species (for example heavy metals, acids, pesticides...)	kg 1,4-DB eq
Freshwater ecotoxicity	Emissions to water and air of substances that damage the ecosystems (flora and fauna) in fresh water (for example heavy metals, acids, pesticides...)	kg 1,4-DB eq
Marine ecotoxicity	Emissions to water and air of substances that damage the ecosystems (flora and fauna) in marine water (for example heavy metals, acids, pesticides...)	kg 1,4-DB eq
Agricultural land occupation	The occupation by men of a certain area of agricultural land for a certain period for agriculture and the landscape changes or space resulting	m ² .year
Urban land occupation	The occupation by men of a certain area of agricultural land for a certain period for agriculture and the landscape changes or space resulting	m ² .year
Natural land transformation	Transformation of a certain area of land	m ²
Metal depletion	Environmental depletion of mineral resources. The calculation performed is based on the remaining stocks and on the current consumption rates	kg Fe eq
Fossil depletion	Environmental depletion of fossil resources. The calculation performed is based on the remaining stocks and on the current consumption rates	kg Sb eq

Table 4
Endpoint damage categories used for the ReCiPe 1.10 calculation method.

Endpoint damage category	Midpoint impact categories considered	Unit
Human health	Climate change Human health, Ozone depletion, Human toxicity, Photochemical oxidant formation, Particulate matter formation, Ionising radiation	DALY
Ecosystems	Climate change Ecosystems, Terrestrial acidification, Freshwater eutrophication, Terrestrial ecotoxicity, Freshwater ecotoxicity, Marine ecotoxicity, Agricultural land occupation, Urban land occupation, Natural land transformation	species. year
Resources	Metal depletion, Fossil depletion	\$

correlation parameter, U₅ the uncertainty factor related to the technological correlation parameter, U₆ the uncertainty factor related to sample size parameter and U_b the basic uncertainty factor (depending on the emissions modelling).

After determining the variance, the statistical analysis of Monte Carlo can be used. This technique consists into running repeated analyses with random input values (over 1000 iterations) to produce a probability distribution. Monte Carlo method helps determining the overall uncertainty of the final results and thus determining how a difference between several scenarios is significant.

Table 5
Linear shrinkage (LS) and loss of ignition (LoI) of the bricks with biobased additives (Ref. being the reference sample without additive).

Sample	Ref.	WS	OSF	GC	DMC
LS (%)	7.7 ± 0.2	6.8 ± 0.1	6.3 ± 0.2	6.9 ± 0.2	8.1 ± 0.3
LoI (%)	9.1 ± 0.1	9.6 ± 0.2	11.2 ± 0.1	11.0 ± 0.1	10.0 ± 0.1

2.2.3. Impact assessment

Impact evaluations were made using the ReCiPe 1.10 calculation method for the 17 midpoint impact categories (Table 3) and the 3 endpoint damage categories (Table 4).

The inventory and the impact calculations were performed on SimaPro 8.0 software.

3. Results and discussion

3.1. Bricks characterization

Several properties of the developed bricks, with vegetable additive (wheat straw or olive stone flour) or biobased chemicals (glycerol carbonate or dimethyl carbonate), were measured and compared to the reference without additive.

The linear shrinkage (LS) and the loss on ignition (LoI) representing the change in dimensions and in mass of the sample during the process are reported in Table 5.

Regarding the incorporation of agricultural additives in the formulations, a decrease of the linear shrinkage can be noticed (7.7% for the reference against 6.3 and 6.8% for the porous ones). This decrease corresponds to a reduction of the contraction of the material. Indeed, the added biomass acts like an inert in the matrix thus contributing to reduce the interaction between the clay particles and consequently causing a reduction of the linear shrinkage [29].

In the case of the incorporation of chemicals, two different behaviors were observed depending on the molecule used. The addition of glycerol carbonate results in a decreased shrinkage

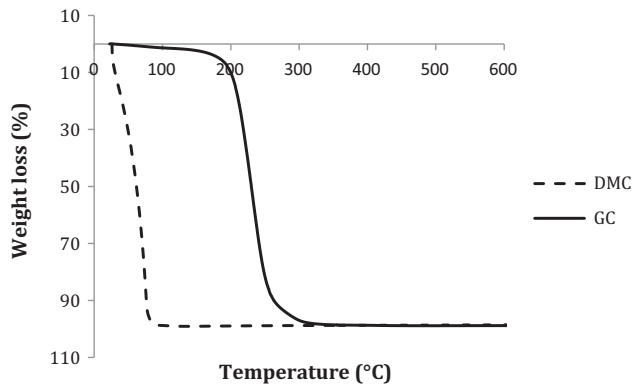


Fig. 4. Thermal behavior of carbonate solutions – determined by thermogravimetric analysis.

while the one of dimethyl carbonate leads to an increase of this value compared to the reference without additive. This could be explained by the apparent difference of viscosity between the two synthetic adjuvants, as well as the needed amount of water to prepare clay samples with the right plasticity [13]. As the linear shrinkage characterizes the evaporation of water out of the matrix during drying, it justifies the highest value obtained for the formulation with dimethyl carbonate, that contains the highest quantity of water (16.2 wt.%).

Regarding the loss of ignition (Table 5), the incorporation of agricultural or synthetic additives leads to products with a higher ignition loss than the reference with only clay (9.1% for Ref. against 9.6–11.2% for the porous samples). Indeed, whether the vegetable matters or the carbonated solutions, they decompose under the effect of the temperature leading to the formation of pores in the matrix.

The thermogravimetric analysis performed on glycerol carbonate and dimethyl carbonate, presented in Fig. 4, shows a complete

decomposition of carbonates from 90 °C for dimethyl carbonate and from 300 °C for glycerol carbonate.

The incorporation of additives also causes changes on other physical properties of the bricks such as porosity and bulk density, as presented in Table 6.

By comparing with the reference value, it can be noticed that the porosity increases with the incorporation of by products: a value of 29.2% is obtained for the sample Ref. against a value of 30.0% for GC and 33.0% for OSF. Meanwhile bulk density decreases from 1.90 g/cm³ to values between 1.79 g/cm³ and 1.88 g/cm³ for respectively the samples OSF and DMC. The incorporation of additives, either vegetable or chemical ones, in the matrix thus leads to an increase of the porosity correlated to a decrease of the bulk density of the samples, as observed in the works of Chiang et al. [30], Russ et al. [31] and Saiah et al. [29] for instance.

The effect of the incorporation rate of the additive may also be highlighted. Indeed, the porosity is higher for the formulation OSF where olive stone flour is added in the amount of 2 wt.%, in comparison to the formulation WS, where wheat straw is incorporated in the amount of 1 wt.%. This increase of porosity is directly related to a decrease of bulk density, with a value of 1.79 g/cm³ for OSF lower than the one obtained for WS (1.86 g/cm³).

As regards the addition of carbonate solutions, the obtained results are quite similar. The porosity of DMC is slightly higher than for GC, while its bulk density is a little more important, which seems quite contradictory. It can be assumed that DMC presents a porosity that was not achieved when measuring the open porosity by immersing the sample into water. This porosity could be either a closed one or one with very small pores, which both could be determined by mercury porosimetry.

Generally, it can be noticed that the impact of the addition of vegetable particles is more important according to the porosity than for synthetic adjuvants. This can probably be explained by the size of the pores formed during the decomposition of these additives, as presented through Scanning Electron Microscopy pictures (Fig. 5).

Table 6
Physical properties of the bricks with biobased additives (Ref. being the reference sample without additive).

Sample	Ref.	WS	OSF	GC	DMC
Porosity (%)	29.2 ± 0.3	31.3 ± 0.3	33.0 ± 0.3	30.0 ± 0.3	30.5 ± 0.1
Density (g/cm ³)	1.90 ± 0.02	1.86 ± 0.02	1.79 ± 0.02	1.83 ± 0.01	1.88 ± 0.01

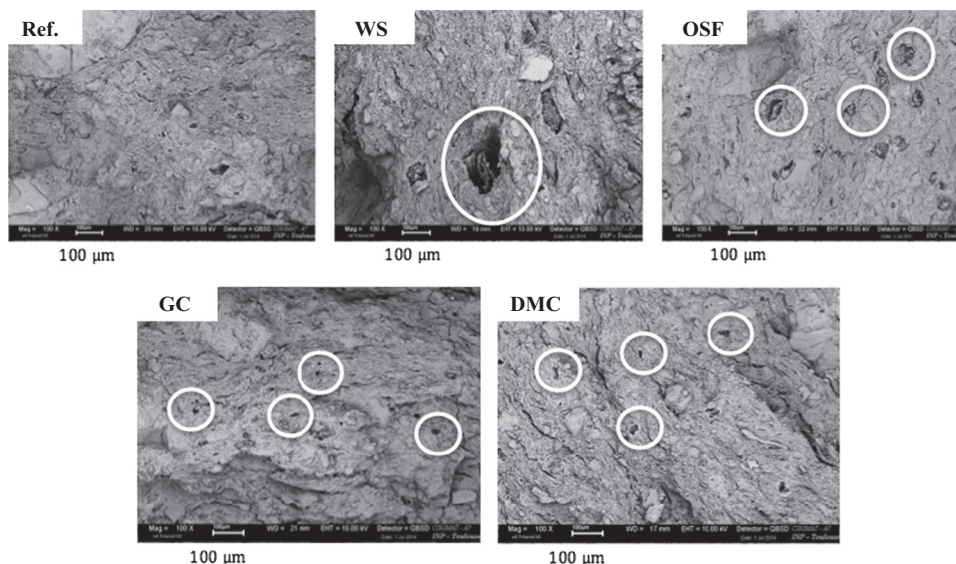


Fig. 5. Scanning Electron Microscopy observations of the porous bricks with biobased additives.

Table 7
Mechanical and thermal properties of the bricks with biobased additives (Ref. being the reference sample without additive).

Sample	Ref.	WS	OSF	GC	DMC
Bending (MPa)	10.4 ± 0.8	10.0 ± 0.8	6.5 ± 0.7	7.0 ± 0.5	7.5 ± 0.8
ThC (W/m.K)	0.57 ± 0.01	0.53 ± 0.01	0.46 ± 0.02	0.53 ± 0.03	0.47 ± 0.04

These shots confirm the formation of pores in all the developed bricks. The pores are of various sizes depending on the adjuvant used, about 100 µm for the formulation WS, 50 µm for OSF and 10 µm for the formulations with carbonate solutions. Moreover, the holes left in the clay matrix have the same shape of the particles before the drying and firing steps: elongated for WS, spherical for OSF and relatively small with irregular shape for GC and DMC.

By producing porous clay bricks, the major aim of such experiments is to develop a material with high thermal and mechanical performances. Thus the thermal and mechanical properties were determined (Table 7).

Concerning the mechanical results, a decrease in the bending strength can be observed for all samples, containing pore forming agents, which is completely logical as increasing the porosity of these materials results in increasing their fragility [32]. In the case of wheat straw, the decrease was not as relevant as the one of the porosity. This could be potentially explained by the densification of the material during sintering, due to new crystalline phases creation (such as unreacted MgO), as presented in Aouba et al. [33] and Njeumen Nkayem et al. [34] works.

In terms of thermal properties, the thermal conductivity was also decreased for all porous samples with values between 0.46 W/m.K and 0.53 W/m.K, compared with 0.57 W/m.K for the reference (without additives), representing a decrease of 7-19% conductivity loss. Indeed, thermal conductivity is mostly influenced by the formation of pores and so the bulk density of the material [35]. But bricks thermal properties do not depend exclusively on their density: other parameters have to be taken into account such as mineral composition, microstructure, humidity or even the nature of the pore system (size, distribution and interconnections) [36-39]. This could potentially explain the result obtained in the case of the addition of wheat straw, where the thermal conductivity decrease is not as important as its porosity increase.

In our study, the objective of obtaining more insulating bricks could be reached by this type of formulation. However, it remains

important to note that the formulations the more insulating also present the lowest bending strengths, as for example the sample OSF with a bending strength of 6.5 MPa and a thermal conductivity of 0.47 W/m.K.

Compromises between mechanical and thermal performances should thus always be performed. In these conditions, the best choice seems to be the incorporation of 1 wt.% of wheat straw (WS), with the best compromise between insulating capacity and mechanical strength. In addition, this sample presents an interesting porosity (31.3%) and a density of 1.86 g/cm³ contributing to lightweight the final brick.

3.2. Life Cycle Assessment of the developed tablets

After characterizing the porous bricks with biobased additives (vegetable and chemical ones) and measuring the insulating capacity of each sample, the aim of this part is to compare the environmental impacts of the five developed formulations. The functional unit has thus been defined as the production of 1 m² of porous tablets with a thermal resistance of 3.5 × 10⁻² m².K/W, corresponding to the one of the reference sample, without pore forming agent.

3.2.1. General comparison of the different scenarios

The general comparison of the scenarios represents the relative percentage of the scenarios on each impact category, the most impacting scenario of the category representing 100% and the others are calculated according to the latter.

The comparison of the different scenarios (Ref., WS, OSF, GC and DMC), using the ReCiPe 1.10 method, is presented in Fig. 6 for the impact characterization and in Fig. 7 for the damage characterization.

The Ref. scenario, without pore forming agent, presents the maximum impact on the 17 impact categories and thus on the three categories of damages (human health, ecosystems and resources) with a gap to the other scenarios between 6 and 22%.

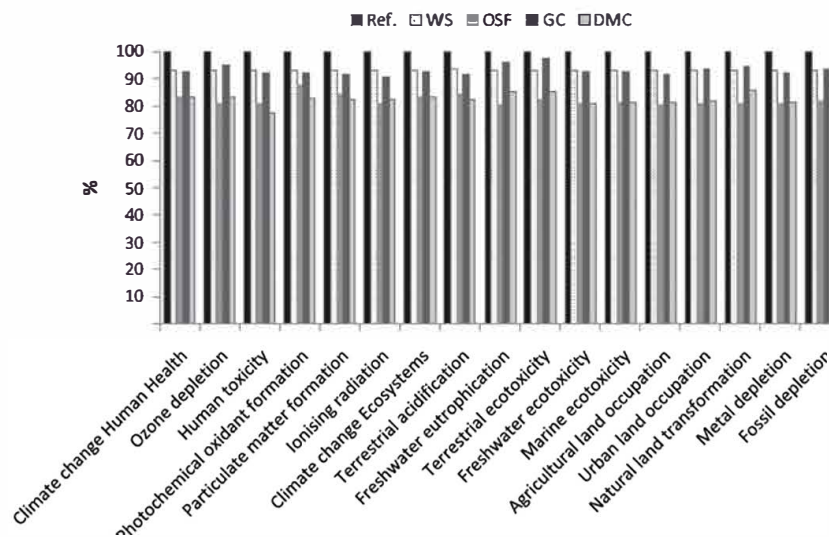


Fig. 6. Comparison of the scenarios with an equivalent thermal resistance – Impact characterization with ReCiPe 1.10 Midpoint (H).

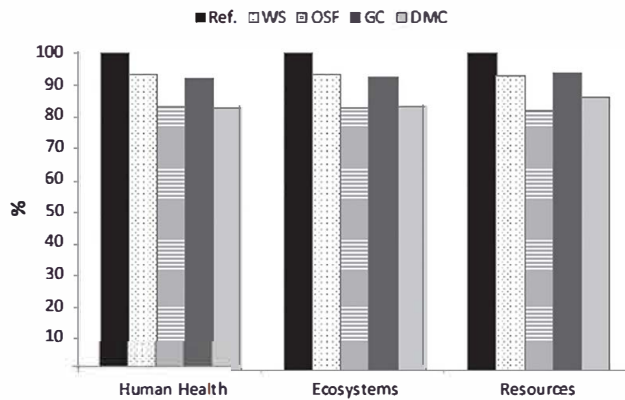


Fig. 7. Comparison of the scenarios with an equivalent thermal resistance – Damage characterization with ReCiPe 1.10 Endpoint (H).

Incorporating biobased additives, whether from agricultural resources or chemical ones, contributes to improve the environmental impacts, for all the midpoint or endpoint categories. In this study, the OSF scenario with incorporation of 2 wt.% of olive stone flour, seems to be the one with the lowest impacts on the environment.

In order to validate the conclusions of this comparison, an uncertainty analysis through Monte Carlo analysis has been realized (Fig. 8). The different scenarios WS, OSF, GC and DMC were compared to the Ref. scenario.

The comparison of scenarios in pairs revealed that the Ref. scenario seems to have a higher probability of occurrence than the others (WS, OSF, GC and DMC), that is to say that there is more chance that the Ref. scenario creates more impacts on the environment than the others.

The difference between these probabilities is significant for the comparisons between Ref. and OSF and Ref. and DMC: all the midpoint impacts present a significant difference from one scenario to another: between 81% and 91% for the comparison Ref. OSF and between 88% and 98% for the comparison Ref. DMC.

On the contrary, for the two other scenarios, the probabilities of occurrence are relatively close to the one of the Ref. scenario. Indeed, by comparing the Ref. and WS scenarios, there is about 65% chance that Ref. is predominant over WS. The same conclusion is noticed for the comparison between Ref. and GC scenarios where Ref. has a probability of occurrence between 56% and 78%.

This uncertainty analysis validates the results presented in Figs. 6 and 7: the incorporation of pore forming agents, either vegetable or chemical ones, contributes to a reduction of the impact on the environment for the developed porous fired clay brick. To our knowledge, no similar studies have been presented yet in the literature.

3.2.2. Hot spot identification: case study of the Wheat Straw (WS) scenario

In an eco design approach, it was envisaged to focus on the scenario presenting the most interesting properties from a

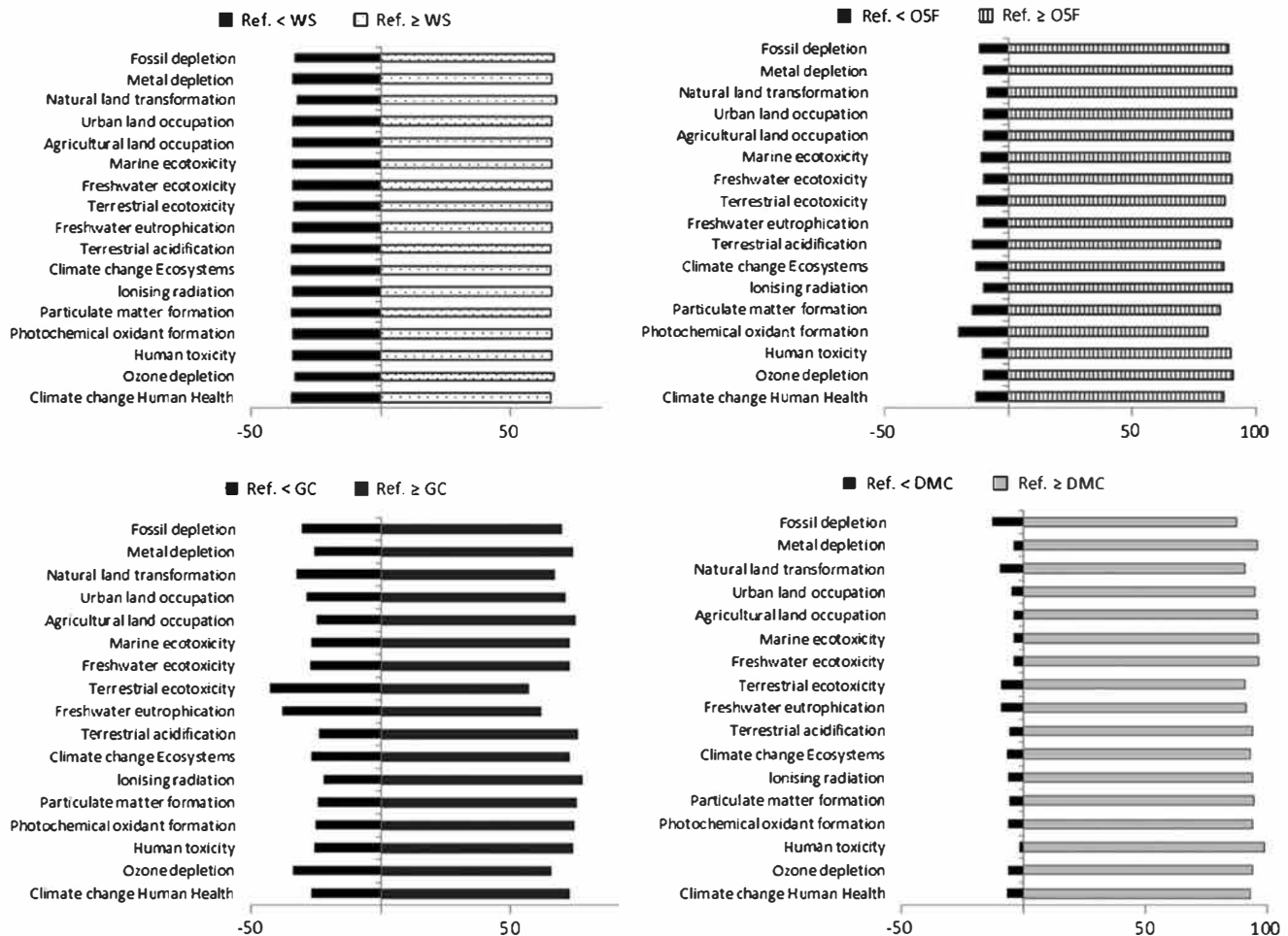


Fig. 8. Monte-Carlo analysis: comparison of all the scenarios with the reference one, without pore-forming agent – Impact characterization with ReCiPe 1.10 Midpoint (H).

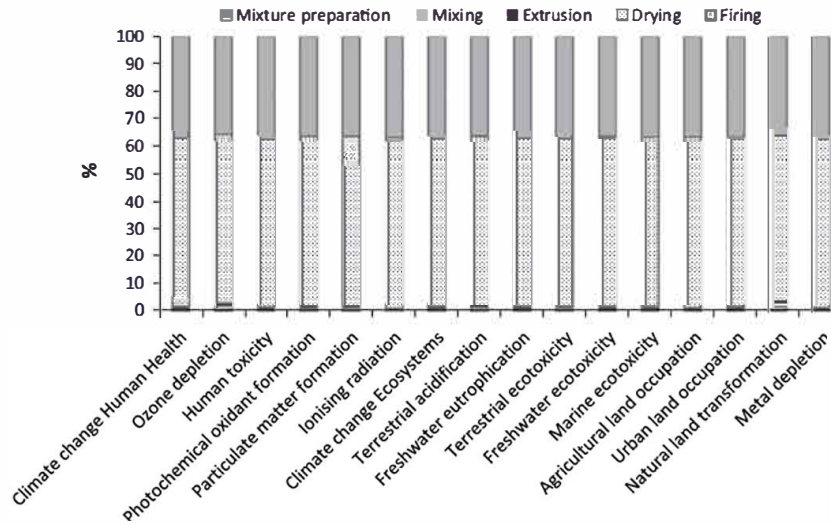


Fig. 9. Hot spot identification of the WS scenario – Impact characterization with ReCiPe 1.10 Midpoint (H).

technical point of view, as presented in Table 7: the WS scenario (with the best compromise between thermal and mechanical performances).

The different steps of the process were then studied one by one to identify those responsible for the majority of impacts. The results are presented in Fig. 9.

It appears clearly that the drying and firing steps are those with the most significant impacts of the life cycle of the products, representing between 94% and 99% for the 17 categories of impacts. Similar results were observed in the literature [7–9]. These results can be explained by the large electricity consumption during these two time consuming steps (respectively 72 h and 12 h for the drying and firing), much larger than for the mixture preparation, mixing or even extrusion stage. These steps being identical for all the 5 scenarios studied, the other modifications (incorporation rate of the pore forming agent or quantity of water added) have not significant impact.

4. Conclusion

In a context of sustainable development and with the new thermal regulations, it is necessary to develop new construction materials with high thermal and mechanical performance. The incorporation of by products has thus been envisaged, also allowing waste recovery combined with a reduction of the associated pollution. In this work, pore forming agents, from agricultural (wheat straw and olive stone flour) or chemical origins (glycerol carbonate and dimethyl carbonate), have been added to the typical clay formulation in order to enhance porosity formation. This increase of porosity has been directly linked to the increase of thermal insulation, whereas mechanical performance collapsed. A compromise had thus to be made in order to develop the best porous fired clay brick with both high thermal and mechanical properties. In our case, the best results were obtained for the incorporation of 1 wt.% of wheat straw, leading to an increase of 7.2% of the porosity, and decreases of 3.8% for the bending strength and 7.0% for the thermal conductivity.

After having developed and characterized new fired clay materials from a technical point of view, a study of the environmental impacts of these five formulations was conducted through a Life Cycle Assessment (LCA), using the ReCiPe characterization method. According to the functional unit selected based on an equivalent thermal resistance, it was noticed that the incorporation of

additives in the matrix, vegetable or synthetic ones, leads to a decrease of the impacts from 15% to 20% (on all the 17 midpoint impact categories and on the 3 damage categories) by comparison with the Ref. scenario, corresponding to the brick formulation without pore forming agent. A clear improvement of the environmental impact of the fired clay bricks is thus possible.

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References

- [1] M. Kommann, Clay building materials: Manufacturing and properties [In French] (Septima), 2005.
- [2] L. Barbieri, F. Andreola, I. Lancellotti, R. Taurino, Management of agricultural biomass wastes: preliminary study on characterization and valorisation in clay matrix bricks, *Waste Manage.* 33 (2013) 2307–2315.
- [3] C. Bories, M.-E. Borredon, E. Vedrenne, G. Vilarem, Development of eco-friendly porous fired clay bricks using pore-forming agents: a review, *J. Environ. Manage.* 143 (2014) 186–196.
- [4] M.S. Mohammed, A.E.-S.I. Ahmed, R.M. Osman, I. Khattab, Combinations of organic and inorganic wastes for brick production, *Polym. Compos.* 35 (2014) 174–179.
- [5] UNEP, Buildings and Climate Change: Status, Challenges and Opportunities (United Nations Environment Programme), 2006.
- [6] L.F. Cabeza, L. Rincón, V. Vilarinho, G. Pérez, A. Castell, Life cycle assessment (LCA) and life cycle energy analysis (LCEA) of buildings and the building sector: a review, *Renew. Sustain. Energy Rev.* 29 (2014) 394–416.
- [7] S. Kumbhar, N. Kulkarni, A.B. Rao, B. Rao, Environmental life cycle assessment of traditional bricks in Western Maharashtra, India, *Energy Procedia* 54 (2014) 260–269.
- [8] H.W. Kua, S. Kamath, An attributional and consequential life cycle assessment of substituting concrete with bricks, *J. Cleaner Prod.* 81 (2014) 190–200.
- [9] C. Koroneos, A. Dompros, Environmental assessment of brick production in Greece, *Build. Environ.* 42 (2007) 2114–2123.
- [10] G.J. Venta, P. Eng, Life Cycle Analysis of Brick and Mortar Products, Sustainable Materials Institute, Athena, 1998.
- [11] Wienerberger, Environmental and Health Declaration Sheets – Porotherm R37 [In French], 2010.
- [12] Imerys, Environmental and Health Declaration Sheets – Monomur 37,5 narrow-joint in fired clay [In French], 2011.
- [13] C. Bories, L. Aouba, E. Vedrenne, G. Vilarem, Fired clay bricks using agricultural biomass wastes: Study and characterization, *Constr. Build. Mater.* 91 (2015) 158–163.
- [14] Y. Fu, H. Zhu, J. Shen, Thermal decomposition of dimethoxymethane and dimethyl carbonate catalyzed by solid acids and bases, *Thermochim. Acta* 434 (2005) 88–92.

- [15] C. Bories, Study of the characteristics of a biobased pore-forming agent and mechanisms used to obtain a micro-porous building brick with high thermal and mechanical properties [In French]. Institut National Polytechnique, Toulouse, Sciences des Agrossources, 2015.
- [16] American Society for Testing and Materials (ASTM), Standard C210-95, Standard Test Method for Reheat Change of Insulating Firebrick, 1995.
- [17] H. Hornain, GranDuBé: quantities related to the sustainability of concrete [In French] (Presses des Ponts), 2007.
- [18] American Society for Testing and Materials (ASTM), Standard C518, Standard Test Method for Steady-State Thermal Transmission Properties by Means of the Heat Flow Meter Apparatus, 2004.
- [19] ISO 8301, Thermal insulation – Determination of thermal resistance and related properties under steady – flow meter method [In French], 1991.
- [20] NF EN 12667, Thermal performance of materials and products for buildings – Determination of thermal resistance by means of guarded hot plate and heat flow meter methods – Products of high and medium thermal resistance. [In French], 2001.
- [21] ISO 14040, Environmental management – Life cycle assessment – Principles and framework. [In French], 2006.
- [22] ISO 14044, Environmental management – Life cycle assessment – Requirements and guidelines. [In French], 2006.
- [23] N. Pargana, M.D. Pinheiro, J.D. Silvestre, J. de Brito, Comparative environmental life cycle assessment of thermal insulation materials of buildings, *Energy Build.* 82 (2014) 466–481.
- [24] A. Nefzaoui, Study of the use of olive by-products in animal feed in Tunisia. [In French] (Tisserand, Alibés), 1983.
- [25] M. Avraamides, D. Fatta, Resource consumption and emissions from olive oil production: a life cycle inventory case study in Cyprus, *J. Cleaner Prod.* 16 (2008) 809–821.
- [26] S. Claude, Z. Mouloungui, J.W. Yoo, A.E. Gaset, Process for the preparation of glycerol carbonate [In French]. Patent FR 2778182, 1999.
- [27] C. Murugan, H.C. Bajaj, Transesterification of propylene carbonate with methanol using Mg-AlCO₃ hydrotalcite as solid base catalyst, *Indian J. Chem.* 49A (2010) 1182–1188.
- [28] B.P. Weidema, M.S. Wesnaes, Data quality management for life cycle inventories – an example of using data quality indicators, *J. Cleaner Prod.* 4 (1996) 167–174.
- [29] R. Saiah, B. Perrin, L. Rigal, Improvement of thermal properties of fired clays by introduction of vegetable matter, *J. Building Phys.* 34 (2010) 124–142.
- [30] K.-Y. Chiang, P.-H. Chou, C.-R. Hua, K.-L. Chien, C. Cheeseman, Lightweight bricks manufactured from water treatment sludge and rice husks, *J. Hazard. Mater.* 171 (2009) 76–82.
- [31] W. Russ, H. Mörtel, R. Meyer-Pittroff, Application of spent grains to increase porosity in bricks, *Constr. Build. Mater.* 19 (2005) 117–126.
- [32] M. Sutcu, S. Akkurt, The use of recycled paper processing residues in making porous brick with reduced thermal conductivity, *Ceram. Int.* 35 (2009) 2625–2631.
- [33] L. Aouba, C. Bories, M. Coutand, B. Perrin, H. Lemerrier, Properties of fired clay bricks with incorporated biomasses: cases of olive stone flour and wheat straw residues, *Constr. Build. Mater.* 102 (Part 1) (2016) 7–13.
- [34] D.E. Njeumen Nkayem, J.A. Mbey, B.B. Kenne Dikko, D. Njopwouo, Preliminary study on the use of corn cob as pore forming agent in lightweight clay bricks: physical and mechanical features, *J. Build. Eng.* 5 (2016) 254–259.
- [35] J. García Ten, M.J. Orts, A. Saburit, G. Silva, Thermal conductivity of traditional ceramics. Part I: Influence of bulk density and firing temperature, *Ceram. Int.* 36 (2010) 1951–1959.
- [36] J. García-Ten, M.J. Orts, A. Saburit, G. Silva, Thermal conductivity of traditional ceramics: Part II: Influence of mineralogical composition, *Ceram. Int.* 36 (2010) 2017–2024.
- [37] M.L. Gualtieri, A.F. Gualtieri, S. Gagliardi, P. Ruffini, R. Ferrari, M. Hanuskova, Thermal conductivity of fired clays: effects of mineralogical and physical properties of the raw materials, *Appl. Clay Sci.* 49 (2010) 269–275.
- [38] E. Rimpel, S.M. El Ghazzali, Influence of the mineral constituents and equilibrium moisture on the thermal conductivity of bricks, in: *Zi-Annual: Annual for the Brick and Tile, Structural Ceramics and Clay Pipe Industries*, Bauverlag, Wiesbaden, pp. 28–52, 1998.
- [39] N.H. Abu-Hamdeh, R.C. Reeder, Soil thermal conductivity: effects of density, moisture, salt concentration, and organic matter, *Soil Sci. Soc. Am. J.* 64 (2000) 1285–1290.