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Ines Labaied, Omar Douzane, Marzouk Lajili, Geoffrey Promis. Bricks Using Clay Mixed with Powder and Ashes from Lignocellulosic Biomass: A Review. *Applied Sciences*, 2022, 12 (20), <10.3390/app122010669>. <hal-03837955>

HAL Id: hal-03837955

<https://u-picardie.hal.science/hal-03837955v1>

Submitted on 5 Dec 2023

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Review

Bricks Using Clay Mixed with Powder and Ashes from Lignocellulosic Biomass: A Review

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Abstract: The production of fired or stabilized bricks from lignocellulosic biomass ash is thoroughly examined in this article. Bricks are typically made through the high-temperature firing process or by stabilizing the mixture with binders such as lime and cement. These bricks have a large carbon footprint and high levels of grey energy. In many parts of the world, the excessive use of clay as a natural raw material for the production of conventional bricks will lead to its scarcity. The mixing of clay with lignocellulosic ash during brick manufacturing leads to a better and more reliable solution that conserves scarce natural resources and reduces the impact of environmental pollution. This study aims to review the state of the art in the production of bricks based on lignocellulosic ashes and their physical, thermal, and mechanical properties. The most recent data in the literature related to the manufacture of lignocellulosic ash-based bricks either by firing, cementing or geopolymerization, the design of mixtures, as well as the identification of the main factors influencing the performance and durability of these bricks are presented and discussed. Despite extensive research, there is still very little commercial use of waste bricks in general and lignocellulosic biomass ash in particular. Various toxicity issues of lignocellulosic ash used in brick production limit their use on an industrial scale due to a lack of appropriate standards. In order to achieve practical production of bricks from lignocellulosic ash, research is still needed on standardizing and sustaining biomass ash recycling.

Keywords: ashes from lignocellulosic biomass; clay; fired bricks; unfired bricks; geopolymer bricks



Citation: Labaied, I.; Douzane, O.; Lajili, M.; Promis, G. Bricks Using Clay Mixed with Powder and Ashes from Lignocellulosic Biomass: A Review. *Appl. Sci.* **2022**, *12*, 10669. <https://doi.org/10.3390/app122010669>

Academic Editors: Chao-Wei Huang, Janusz Lasek and Yueh-Heng Li

Received: 9 September 2022

Accepted: 16 October 2022

Published: 21 October 2022

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1. Introduction

In front of the requirements of comfort and the increase of the real estate parks, one observes that the building and public works sector is currently in whole proliferation. This yields a huge growth in the consumption of natural resources and negatively affects our environment. Indeed, the materials which are now used in construction are energy-consuming, not ecological since they are made of minerals and carbon-based materials that are not biodegradable. Moreover, the production of such materials is always inherent of pollutant emissions gaseous and particulate matter PMs. According to the International Energy Agency (IEA), the building sector alone consumes about 30% of the world's energy supply and is responsible for almost 40% of greenhouse gas emissions [1].

Clay brick is one of the most basic building materials used in the construction field worldwide. Brick factories are very energy-intensive businesses. In 1998, a brick factory with a daily production capacity of 400 tons used up to 5 GWh of electricity and 56 million thermies (1 thermie = 4,185,500 joules) of natural gas annually, for total annual energy consumption of 70 GW [2]. Furthermore, this energy consumption accounts for approximately 50% of the total cost of production. As a result, the consequences are significant for brick manufacturers, who would have to modify their processes. Hence, the reduction of energy consumption and emissions in the manufacture of bricks, especially during the drying and

firing stages, the reduction of production costs, and the preservation of natural resources are nowadays the main challenges of the ceramic industry. These objectives not only enter in the context of sustainable development but also in a context of increased competition with the new promoters of building materials such as prefabricated concrete and cellular concrete which became the dominant materials in the sector of the building. Nevertheless, clay is still the main raw material in the manufacture of bricks, a non-selective and blind consumption of non-renewable sources will lead to the depletion of this natural material. That is why the investigation of alternative bricks using clay and biomass would be a promising path for reducing pollution impact on the environment, improving the characteristics of the brick, and preserving the natural clay resources. Moreover, the use of biomass by-products and wastes in a virtuous circular economy for society is a long-standing issue in some countries [3]. Indeed, recycling is part of the 3Rs waste treatment strategy: Reduce, Reuse, and Recycle [4]. In this context, many reviews focusing on the use and the valuation of biomass wastes in the construction and building field and more specifically in the manufacturing of fired and unfired bricks were reported in the literature. Many studies focused on fired bricks investigated the effects of the addition of waste products and by-products derived from renewable resources, including agricultural residues and mineral resources in the manufacture of fired clay bricks. The researchers claim that using this waste is a useful solution because it is not only capable of acting as pore-forming agents during the firing process and creating pores in the clay matrix, making bricks lighter and more thermally insulating, but also can be valorized and recycled as toxic wastes that cause environmental harm. However, the use of these wastes in large quantities is detrimental to the mechanical properties of the fired bricks, so it is crucial to find a compromise between thermal and mechanical properties to achieve an innovative product. Ref. [5] provide a review of the effects of the chemical composition of clay, as well as the various types of additives that are used (agro and non-agricultural wastes), the manufacturing procedures in comparison to the factory scale, and the effects of adding these types of wastes on the clay bricks' compressive strength, density, and water absorption. Despite the difference in the percentages of waste used, as well as the firing temperature in the different studies carried out, Ref. [5] found an increase in absorption, thermal resistance, and a decrease in compressive strength by increasing the percentage of waste used in the manufacture of fired bricks. The effect of flux oxides from wastes on shaping technique (extrusion, pressing, and molding) and brick production (molding pressure, soaking time, and firing temperature) as well as on fired clay brick physical and mechanical properties were studied by [6]. As a result of the heat treatment, the heavy metals contained in these wastes stabilize in the clay silicate structures, thus reducing their leaching potential. However, much of the research attention has been concentrated on unfired clay bricks [7–11]. This is mainly because of the environmentally friendly manufacturing process and secondly because of the relatively low energy consumption. Ref. [8] studied the stabilization effect of waste, mainly ash, industrial waste, and mineral waste by cement binders (lime/cement) or chemical binders in the manufacture of fired bricks. They stated that strategy, time, speed, and material by mixing, method of compaction, method of curing, and type and quantity of binder are key parameters for obtaining good physical and mechanical quality of the final product elaborated. It is important to note that recent developments in the unfired brick industry are oriented toward geopolymer bricks. Against this background, Ref. [7] presented an examination of the development of geopolymer bricks by alkaline activation of industrial waste. Compared to conventional bricks, alkali-activated geopolymer bricks have been found to be stronger, more durable, and more environmentally friendly, especially since they are produced with a high waste content without the addition of a cementitious binder. In addition, Refs. [9,10] were interested in fired and unfired bricks reinforced by different wastes and materials and they particularly focused on their physical, thermal, and mechanical properties.

However, the primary goal of the current review paper is the valorization of lignocellulosic biomass ash when used as a substitute material for the production of fired and unfired

bricks. A significant amount of this waste is produced annually by the burning of various types and forms of solid biofuels that are fed into boilers, stoves, and furnaces [11,12]. Incorporating ash into ceramic production would be a promising way to lessen the dangers associated with this kind of waste's impact on the environment, ease the strain on clay resources, and cut down on energy use and carbon dioxide emissions. Therefore, one tries to give a thorough overview of the various raw materials used, the various processes of making fired and unfired bricks, and the various physical, thermal, and mechanical properties. All this was carried out without neglecting the environmental positive impact when investigating using the ash during the production of new bricks to facilitate their integration into the building material markets.

2. Composition of Lignocellulosic Biomass and Clays Properties

2.1. Structure and Texture of Lignocellulosic Biomass

According to the European Directive 2003/30/EC, biomass is all types of biodegradable products/by-products from forestry residues, woody processing industries, agriculture (animal and plant residues) as well as municipal and industrial waste (food industries), allowed for energy use in accordance with environmental protection regulations [13]. Hence, biomass can be classified into four types: Forestry waste, Agricultural waste, Aquatic residues, and other origins. The waste of agricultural origin and forestry are classified as lignocellulosic biomasses. Lignocellulosic biomass is a fibrillar biological structure composed of cellulose, hemicellulose, and lignin as it is illustrated in Figure 1, in relatively small proportions of non-nitrogenous extractives, crude proteinaceous material, lipid, and mineral matter. Plant species, age, and organs determine the proportions of these constituents [14].

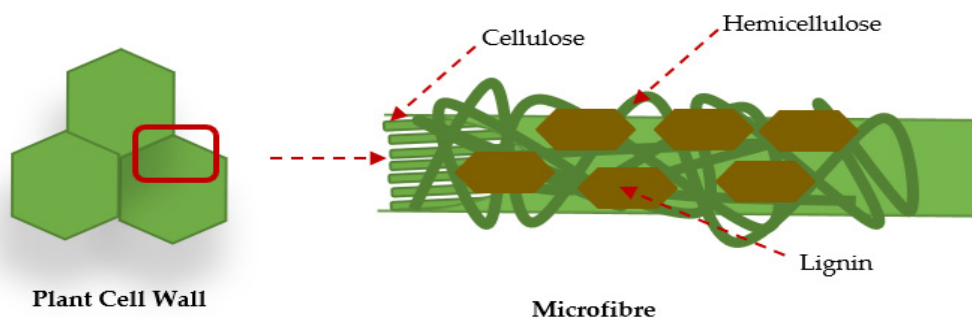


Figure 1. The main structure of lignocellulosic biomass.

The world's production of lignocellulosic biomass is estimated at more than 220 billion tons per year [15]. In order to valorize this type of waste and preserve the environment, biomass can be used in animal feed, soil fertilization, as well as biofuel. Indeed, lignocellulosic biomass has a significant calorific value thanks to its chemical composition rich in organic matter and combustible carbon, which favors its energy valorization as biofuels [16]. Currently, biomass contributes between 8% and 15% of the world's energy supply in the form of heat and/or electricity and transport fuels. Therefore, around 476 million tons of biomass ash are generated worldwide each year through the biomass burned is estimated at 7 billion tons with an average ash yield of 6.8% on a dry basis [17]. These ashes are classified as hazardous materials because they contain high percentages of heavy metals. It has been proved that the addition of such by-products (olive waste, rice husk, palm waste, paper pulp residues, *Typha Australis*, sunflower seed cake, wheat, and sawdust straw . . .) constituted an innovative and effective way of generating more pores in the fired bricks [18–26]. It was shown that a small quantity of additives (usually less than 10% by weight), which burns during the firing process, leads to an increase in the porosity of the brick and thus to a decrease in thermal conductivity. However, it was also observed that this creation of pores generally led to a significant decrease in mechanical performance.

2.2. Natural Clays as the Main Component in Construction Materials Such as Bricks

Clay is a natural mineral product, abundant and of small granulometry that, in the presence of water, forms a plastic paste that hardens after firing. These properties have attracted greater attention of researchers as a primary source in the ceramic industry and more specifically in the manufacture of bricks. Moreover, different factors can affect the quality, strength, and durability of these produced bricks, including particle size, Atterberg limits, calcium carbonate content, and chemical composition.

2.2.1. Particle Size Analysis

The clay granulometry gives orientations on the quality of the elaborated bricks. Indeed, to obtain good quality bricks, the soil must not be too clayey so that the clay fraction of the soil ($<2 \mu\text{m}$) must be higher than 5% and lower than 30% playing the role of a binder and the coarse grains (silt and sand) must have an average diameter lower than 5 mm giving the stability of skeleton [27,28] highlighted the importance of clay granulometry in the manufacture of compressed earth bricks stabilized by residual calcium carbide (CCR) and rice hull ash (RHA). He tested the effect of granulometry on the microstructure and mechanical strength of earth bricks reporting the percentages and diameters of each particle size family. However, most of the research work on new forms of bricks made from biomass waste and clays with a particle size between 150 and 600 μm has not provided any precision either on the percentages of clay fraction or on the diameter of the sand and silt fraction. Nevertheless, the granulometry of the clay was crucial since it allows us to judge useful or not the use of the sand as a degreaser, and it provides also information on the plasticity of produced samples [29]. Indeed, the finer the grains, the more plastic the substance. In addition, the smaller the particles, the more reactive they are and promote rapid densification.

2.2.2. Atterberg Limits

The Atterberg limits are geotechnical parameters intended to identify soil and determine its consistency limits. They allow an approximate value on the quantity of water necessary during the shaping of bricks. In addition, they provide essential information on the mechanics of the soil via the determination of its plasticity index. For instance, it is recommended that the plasticity index of the soil should be between 5% and 15% for bricks because the highly plastic nature causes the excessive use of water. However, after the drying and firing phases, shrinkage can cause cracks and other defects that decrease the compressive strength of the final product. [30–32], have determined the Atterberg limits such as liquid limit, plastic limit, and plasticity index of the clay blended with 0%, 2%, 4%, 6%, 8%, and 10% of rice husk ash (RHA), respectively. It was shown that there was no relationship between yield strength and RHA, but the plasticity limit decreases with increasing RHA content. This may be due to the reduction of the amount of free silt and clay fraction by forming coarser materials with larger surface areas that require more water. Furthermore, it was reported that the incorporation of up to 10% of RHA waste could be acceptable in the manufacture of bricks without losing plastic behavior. Nevertheless, further studies are recommended when investigating the effect of soil plasticity on the strength properties of bricks using lignocellulosic biomass ash.

2.2.3. Chemical Composition

Physical and mechanical properties of bricks are strongly influenced by the composition of the clay [33]. Silica (SiO_2), the main component of clay, exhibits the highest content compared to other elements which allows it to play an important role during the skeleton brick elaboration. However, silica content higher than 80% increases the porosity and thereby the risk of cracking during the cooling stage [34]. The second main component of the clay is alumina (Al_2O_3). It influences significantly the plasticity of the clay which improves the compressive strength [5]. In addition, the presence of iron oxide Fe_2O_3 is important in terms of retention of metallic trace elements and in terms of coloration of

the fired products. Indeed, the higher its content ($\geq 5\%$) is, the more reddish the coloring becomes. However, for values ranging between 1% and 5%, the bricks were characterized by light brown color. Moreover, the presence of Fe_2O_3 , with a content $>10\%$ causes mold problems, especially when the clay is homogenized for very long periods or in the case of firing in kilns was poor in oxygen. Calcium carbonate CaCO_3 presents an important effect on the mechanical strength of the brick. This phenomenon could be related to the release of CO_2 and CaO during the firing process. Moreover, the release of CaO is associated with the SiO_2 present in the clay which improves the mechanical performance of the fired bricks. Values $>18\%$ favor the presence of free lime which causes cracks and expansions due to excessive moisture absorption [5]. Minerals such as K_2O , MgO , Na_2O , and CaO can promote the formation of glassy phases at lower temperatures as melting oxides. Their proportions affect the firing temperature, which is lower when they present high contents [35].

3. Different Types of Ordinary Bricks: Preparation and Characteristics

3.1. Ordinary Fired and Unfired Bricks Preparations

The history of brick-making dates back more than 7000 years ago (for example, Çatalhöyük was a very large Neolithic and Chalcolithic proto-city settlement in southern Anatolia, which existed from approximately 7500 BC to 6400 BC, and flourished around 7000 BC) when bricks were made in the form of hand-molded earth blocks without compaction and dried in the sun [9]. In order to meet both the growing demand for brick consumption and the new standards of comfort in the building, many techniques were considered in order to improve the manufacturing processes of bricks. The mixing process is carried out with different sorts of industrial mixers allowing homogeneous mixtures. The obtained mixture should be compacted in order to ensure a uniform density and the compactness of the mixture for improving the mechanical performances of the elaborated bricks [36]. Different techniques could be considered such as; compaction, forced compaction, self-compaction, or manual compaction. However, as shown in Table 1 the majority of reported papers used axial compression using a hydraulic press with only precession of the compaction pressure. Moreover, different curing conditions affect the quality and performance of the bricks, especially the environmental conditions such as temperature, humidity, and pressure, as well as the curing time and medium. It is to be highlighted that unfired bricks can be cured by several methods such as:

- Air/kiln curing: this technique can be realized at room temperature with relative humidity similar to that of the natural environment (the reason for the air curing name) or in laboratory ovens at a temperature ranging between $35\text{ }^\circ\text{C}$ and $115\text{ }^\circ\text{C}$ (named kiln curing in this case) [37]. For air-cured bricks, and in order to preserve the molding moisture and vaporization losses, the bricks could be covered with plastic sheets or placed under wet jute [38].
- Wet curing: it is a curing process with high relative moisture (95–100%), but at ambient temperature [39]. This curing can be carried out in a steam atmosphere (steam curing appellation) with the same relative humidity as the wet curing, but when increasing the temperature between $45\text{ }^\circ\text{C}$ and $75\text{ }^\circ\text{C}$ [40].
- Water curing: consists of immersing the samples in water at ambient or elevated temperatures. To ensure that the sample has adequate green strength, this step should be completed immediately after the wet curing step or after the air curing [41].

Although the manufacture of unfired bricks exhibits several advantages, it also presents some disadvantages; this is mainly the lack of strength, durability, and the limitations of professional rules, respectively. To overcome these challenges and to seek durable bricks with good mechanical performance, today's bricks are primarily produced by stabilization using binders such as lime, cement, chemical binders, or a mixture of them, or by firing. The process of firing in kilns at relatively high temperatures permits the clay, as a ceramic vitrified or semi-vitrified, to be transformed into an amorphous solid devoid of any crystalline structure similar to glass. Clay vitrifies somewhere around $573\text{ }^\circ\text{C}$ corresponding to the inversion of quartz, whereas the temperature range between $1000\text{ }^\circ\text{C}$

and 1050 °C is associated with the fusion of clay [42]. Following the extraction of the clay, the mixture must be prepared including two main steps: crushing in the first, and dosing and combining raw materials in the second. Industrial manufacture generally works by extrusion with the help of an Archimedean auger, but it is possible to use other mechanisms such as hydraulic pistons, rolling mills, or presses. The drying process should continue until the bricks contain only about 2% of water in order to reach optimal plasticity. The firing process is the last step during which the shaped and dried clay brick must undergo before it can definitively become a terra-cotta brick. In tunnel kilns, the temperature should be gradually increased until reaching the firing temperature (between 850 °C and 1200 °C, depending on the type of clay). The temperature should be also gradually decreased until the product is completely cooled.

3.2. Possibilities to Improve the Brick's Characteristics

For many years, the brick industry has been mobilizing to minimize its environmental impact. Using waste ash instead of soil is seen as a viable and ecological option for preserving natural resources and reducing ash toxicity and landfill problems. Researchers have incorporated various ashes in fired and unfired bricks as a replacement for clay such as fly ash [43–45], bottom ash [46,47], round blast furnace slag [48], and incineration ash [49,50]. Compared to other industrial ashes, lignocellulosic biomass combustion ash has not been thoroughly investigated in the production of bricks. For fired bricks, the role of biomass ash changes in the clay matrix during the drying and firing stage depending on the origin and nature of these used co-products. The main roles of these ashes found in the literature are:

- Pore-forming agent: Since biomass ashes are composed of a percentage of organic matter, degradation during the firing stage occurs while creating pores within the clay matrix. In addition, this type of ash is used to decrease the density and thermal conductivity of the bricks for more thermally insulating building materials [6,23,51].
- Fluidizing agent: during firing, the melting oxides in the ash react with the silica and alumina in the clay to form a liquid phase and lower the melting point of the bricks. This contributes to the rapid densification and consolidation of the clay material [52–54].
- Clay substitute or sand: the biomass ashes of inorganic co-products have strong plasticity. They can then play the role of degreaser within the clay matrix, producing a glassy phase during firing [55,56].
- Biomass ashes are waste products rich in organic matter and a percentage of unburned carbon. Their role is to act as a biofuel during the firing stage, reducing the temperature and energy consumption of the drying and firing process of the bricks in the tunnel kilns.

For unfired bricks, research is moving toward using ash-based geopolymer bricks instead of cementitious binders as an ecologically sound, economically sensible, and sustainable binder that uses less energy and exhibits strong properties such as good mechanical properties, low permeability to liquids, resistance to high temperatures and acid attacks, and a significant reduction in CO₂ emissions. Indeed, any powder containing high silica and alumina contents and having an amorphous phase can be considered a geopolymer precursor. It is worth noting to recall that the geo-polymerization process is divided into four main steps: (1) The Dissolution of the oxidized minerals (silica and alumina) present in the raw material in an alkaline solution. (2) The diffusion of the dissolved oxidized minerals from the surface to the inner particular space and followed by coagulation and gel formation. (3) The Polycondensation forming a three-dimensional alumina-silicate network and (4) the hardening of the matrix.

4. Investigation of Hybrid Bricks by Blending Clay with Ash from Lignocellulosic Biomass Combustion

4.1. Different Types of Collected Lignocellulosic Biomass Ash Wastes and Their Chemical Components

Depending on the origin of lignocellulosic biomass and the used equipment, the rate of ashes and their chemical composition varies [57]. There are two main types of ash: bottom ash and fly ash. Bottom ash or under-furnace ash is issued from grates and circulating fluidized beds. However, fly ash is composed of coarse fly ash from the dust collector (multi-cyclone) after the treatment of fumes and fine ash from a bag filter or electro filter. Fly and bottom ashes are mainly composed of silica (SiO_2), phosphates (P- P_2O_5), oxides, hydroxides, carbonates, silicates, sulfates, calcium, potassium, sodium, magnesium, organic matter, total unburned organic carbon and traces of metals such as Cr, Ni, Zn, As, Se, Mo, Cd, Pb, Hg, respectively. In order to avoid the effects of these ashes on the environment, they must be interred in cells designated for hazardous or non-hazardous wastes based on their rates [58]. Since this method is very expensive, several alternatives are investigated such as chemical and thermal stabilization [59]. The chemical stabilization could be produced either by the pozzolanic reaction. This process leads to the precipitation of a variety of hydrated calcium silico-aluminates as binders which become promising and sustainable alternatives in the manufacture of unfired bricks. The following step is the thermal stabilization which consists of mixing these ashes with clay and heating the mixture in high-temperature furnaces.

4.2. Hybrid-Fired Bricks by Blending Clay with Lignocellulosic Biomass Ash

To study the effect of ashes on clay bricks, Table 1 summarizes several works carried out and various parameters considered during clay brick preparation. Ashes from olive wastes were the most commonly used, especially in the Mediterranean region where this ash type is produced in huge quantities. Indeed, several researchers have chosen to use these wastes as partial replacements in the manufacture of clay bricks in order to reduce the environmental effect of these ashes and to decrease the aggressive use of clay as a raw material in the ceramic industry. Ref. [60] used olive bottom ash (OPBA) from the combustion of Spanish olive pomace in a steam boiler mixed with an equal share of three types of clay: yellow, red, and black raw clay, respectively. The replacement rate was ranging from 10% to 50%. As shown in Table 2, the olive bottom ash (OPBA) was formed by 12.5% SiO_2 and Al_2O_3 which represent the skeleton components of bricks, 53.1% of melting oxides (mainly CaO , Na_2O , K_2O , and MgO), and gaseous components formed by 1.6% FeO_3 , 42.28% CaCO_3 and 13.5% of organic matter. The latter is responsible for the generation of gases and swelling of ceramic bodies during sintering at high temperatures. The presence of significant levels of melting oxides (K_2O) and auxiliary melting oxides (CaO , MgO , Na_2O) in the OPBA could lower the melting point which could reduce the firing temperature. This study proves the feasibility of using OPBA up to 20% as a partial substitute for clay to produce good quality of fired bricks. However, only 10% weight of OPBA displays the optimum value that confirms the good balance provided by both melting capacity and porogen capacity. In the same context, Ref. [61] used different percentages (5%, 10%, 15%, 20%, and 25%) of fly ash from the combustion of olive pomace (CVLL) as a partial substitute for clay (black, red, and yellow). These ashes came from the same plant and same source as those used in the earlier study. 1500 tons of bottom ash and 5400 tons of fly ash were produced annually at that plant. These ashes were mainly composed of K_2O , silica, and auxiliary flux oxides (CaO , MgO) (Table 2). Based on experimental results, it was shown that when using fly ash up to 25% by weight in the clay brick, the final product was useful in the manufacture of ceramic bricks for building materials. But, for obtaining bricks with superior physical and mechanical properties, a limitation of 5% by weight of CVLL should be considered. Furthermore, Ref. [62] evaluated the effect of the incorporation of olive stone ash (OSA) as a secondary raw material on clay bricks' physical, mechanical, and thermal properties. A mixture of 30% red clay, 30% yellow clay, and 40%

black clay from Spain were mixed with 10%, 20%, and 30% olive stone ash by weight. As demonstrated in the study, the incorporation of 10% weight of olive stone ash in clay bricks ensured a balance between bulk density, bulk porosity, water absorption, and tensile strength. Ref. [63] incorporated various industrial wastes into clay to produce fired bricks. Sugar bagasse ash (B) and oil mill wastewater (OMW) were used at a 2.5% and 6.5% content, respectively. OMW and B act as porous agents, reducing the thermal conductivity of clay during the firing process. Ash from rice husks and sugarcane bagasse were also frequently used in the production of clay bricks. In fact, Refs. [31,32] used leftover rice husk ash (RHA) to make clay bricks with contents of 0%, 2%, 4%, 6%, 8%, and 10%, respectively. Ref. [64] developed clay bricks with different levels of clay replacement ranging from 0% to 20% with rice husk ash. Ref. [31] showed that 4% by weight was the optimum amount of RHA waste that can be used for improving the structural, thermal, and acoustic characteristics of clay bricks. Ref. [32] showed that the addition of only 2% of RHA to clay was sufficient to improve the mechanical properties of clay bricks. As for [64] 5% of RHA introduced in clay could produce high-quality clay bricks. In addition, Ref. [65] investigated the viability of using waste biomass combustion ash, such as (RHA) or plank wood ash (WA), with contents ranging from 10% to 30%, in the production of clay bricks. Two different firing temperatures, 900 °C and 1000 °C were tested to determine the effect of temperature. According to the results, 10 wt.% RHA and 20 wt.% WA, as well as a firing temperature of 1000 °C, provided the best mechanical, physical, and thermal properties. Ref. [66] investigated the viability of reusing waste sugarcane bagasse ash (K) for use as a secondary raw material during the production of clay-fired bricks. The replacement rate of natural clay was limited to 20% by weight in 5% steps. According to the findings, sugarcane bagasse ash waste (K) can be safely and sustainably used in clay bricks when its weight is less than 10%. Refs. [67,68] and studied the mechanical, physical, and thermal performance of new forms of clay bricks by adding various dosages of 5%, 10%, and 15% of Rice husk ash RHA and sugar bagasse ash (SBA). According to [67], bricks containing up to 5% RHA and SBA can be used in the building and construction industry without causing any issues. Additionally, by adding these ashes, the matrix becomes more porous, making the bricks lighter. However, according to [68], using SBA and RHA up to 15% by weight of clay when making fired clay bricks not only helps to reduce landfill waste but also promotes the development of environmentally friendly and energy-efficient buildings. Palm waste has also begun to be utilized in the ceramic industry. Indeed, Refs. [69,70] studied the feasibility of using palm oil fuel ash (POFA) in the manufacture of fired clay bricks as an alternative to minimize the disposal of this waste in landfills. For [70], 0%, 1%, 5%, 10%, 20% and 30% of POFA waste were introduced in order to design clay bricks fired at 1050 °C. The results show that incorporating this waste up to 5% was effective in producing strong, light, affordable, and environmentally friendly clay bricks. Ref. [69] examined the possibility of incorporating POFA waste with high percentages of 5%, 10%, 15% and 20%, respectively, into clay bricks under real industrial conditions. Contrary to [70], it was proved that the use of OPFA up to 5% as a partial replacement of clay on a large scale is not only useful to minimize the environmental impacts related to the disposal of this waste but also to reduce the excessive use of clay in the ceramic sector.

Table 1. Studies on the production of bricks from lignocellulosic biomass ash waste materials through firing.

Article	Biomass Ash	Clay	Reinforcement Rate	Moisture of Molding	Forming Technique	Drying	Firing	Cooling	Characteristics Examined Bricks
[60]	Bottom ash from olive pomace of combustion (OPBA) (150 μ m)	Equal amounts of Spanish red, yellow, and black clay (150 m)	10–20–30–40–50% of bottom ash	7–10 wt. %	Uniaxial pressure of 54.5 MPa	105 °C for 24 h	Laboratory furnace at a rate of 3 °C/min up to 950 °C for 4 h	Natural convection inside the furnace	Linear shrinkage Water absorption Open porosity Closed porosity Bulk density Compressive strength Development of porosity Thermal conductivity, Leachability
[61]	Fly ash from olive pomace of combustion—(CVLL) (150 μ m)	Equal parts of red, yellow, and black clay from Spain (150 μ m)	5, 10, 15, 20, and 25 wt.% of fly ash.	8% μ	Uniaxial pressure of 54.5 MPa	110 °C for 24 h	Heating rate of 2 °C/min up to 950 °C, for 1 h	Until reaching an ambient temperature	Linear shrinkage Loss on ignition Apparent density Water absorption Compressive strength, porosity
[62]	Olive stone ash (OSA) < 100 μ m	30% red, 30% yellow and 40% black clay from Spain (500 μ m)	10, 20, 30%	-	Uniaxial pressure of 54.5 MPa	-	Laboratory furnace at a rate of 3 °C/min up to 900 °C for 4 h	Cooled to room temperature by natural convection inside the furnace	Loss on ignition Linear shrinkage Bulk density Water absorption Compressive strength Thermal conductivity
[63]	Bagasse ash olive mill (B), wastewater ash (OMW), 150 μ m	Red, white and black clay (C) 150 μ m	2.5 mass% bagasse (B) 6.5 mass% OMW	8%	Uniaxial pressing of 54.5 MPa	48 h at 110 °C	In a laboratory-type electrically heated furnace at a rate of 10 °C/ min to 950 °C for 6 h.	-	Loss on ignition Linear shrinkage Bulk density Water absorption Compressive strength Thermal conductivity
[65]	Rice husk ash (RHA) wood ash from boards (WA) < 100 μ m	30 wt.% red, 30 wt.% yellow, and 40 wt.% black clay (500 μ m)	0–30 wt. %	7–10 wt. %	54.5 MPa of pressure	-	3 °C/min up to 900 °C or 1000 °C for 4 h	Cooled to room temperature by natural convection inside the furnace	Linear shrinkage Water absorption Open porosity Closed porosity Bulk density Compressive strength Thermal conductivity Leachability of heavy metals
[67]	Rice husk ash (RHA), sugarcane bagasse ash (SBA)	Clay from a brick kiln in Pakistan (C)	5%, 10%, 15% of RHA and SBA by weight of clay	-	-	Dried in the sun for 10 days	Burnt in a kiln for 36 h at approximately 800 °C	45 days from the kiln	Bulk density Water absorption Compressive strength Efflorescence

Table 1. Cont.

Article	Biomass Ash	Clay	Reinforcement Rate	Moisture of Molding	Forming Technique	Drying	Firing	Cooling	Characteristics Examined Bricks
[31]	Rice husk ash (RHA), a residual of the brick firing process	Clay from Dankotuwa	2%, 4%, 6%, 8% and 10% by weight of the brick	-	Without applying pressure	Under the warm weather condition (35 °C, and 60% relative humidity)	600 °C and 800 °C in a brick klin	-	Atterberg limit test Water absorption Compressive strength Thermal performance Acoustic performance
[32]	Rice husk ash collected from rice mill in Wadata, Makurdi (RHA)	The soil of EljuleOjeb	2% to 10%	-	15 kN/m	Room temperature for 12 days prior to burning	In an electric muffle furnace rise of 200 °C/h and a 4 h soak at 800 °C.	-	Atterberg limits, specific gravity, compressive strength and water absorption tests
[64]	Rice husk collected from the northern region of Malasia (75 µm)	clay	0% to 20%	50 g	-	-	950 °C for 2 h	-	Water absorption Compressive strength
[69]	Palm oil fuel ash (POFA), from the palm oil industry	Clay from local kiln bricks in Makassar (Indonesia)	0%, 5%, 10%, 15% and 20% by weight of clay	-	Without applying pressure	8 days in sunlight	750–850 °C for 96 h using wood and rice husk as fuel (industrial furnace)	-	Density Water absorption Initial rate of absorption Flexural strength Compressive strength Efflorescence sulfate resistance
[70]	Palm oil fuel ash (POFA) from a palm oil mill (Kluang, Johor) 2.36 mm	Clay soil was from a brick manufacturer (Yong Peng, Joho) 2.36 mm	0%, 1%, 5%, 10%, 20% and 30%	476–493, 524 and 557 mL	Pressure of 2000 ps I (13.78 MPa)	24 h at room temperature, followed by an oven drying period of 24 h at 105 °C	1 °C/min up to 1050 °C	-	Firing shrinkage Dry density Initial rate of absorption water absorption Porosity Compressive strength
[68]	Sugar bagasse ash (SBA) was acquired from a sugar mill whereas. Rice husk ash (RHA) from a local brick industry.	Clay (soil)	5%, 10%, 15%	20.8–22.9%	-	Sun-dried for 2 days	800 °C	In the kiln for 45 days	Compressive strength Apparent porosity Water absorption Thermal conductivity
[66]	Sugarcane bagasse ash (K) waste collected from a sugarcane plant located in south-eastern Brazil	Clay	0, 5, 10, 15 and 20 wt.%	7% moisture dry basis	Uniaxial pressing at 21 MPa	110 °C for 24 h	At 1000 °C in an electrical kiln	-	Linear shrinkage Water absorption Apparent density Tensile strength

Table 2. The chemical make-up of various lignocellulosic biomass ashes used in the production of fired bricks.

Oxide Cotenant	OPBA [60]	CVLL [61]	OSA [62]	OMW [63]	B [63]	RHA [31]	RHA [32]	RHA [64]	WA [65]	RHA [65]	K [66]	RHA [67]	SBA [67]	RHA [68]	SBA [68]	POFA [70]	POFA [69]
SiO ₂	10.88	11.7	8.47	37.25	54.53	84.14	49.30	22.12–90.20	48.60	76.7	61.59	77.21	87.97	75.42	85.41	54.7	67.74
Al ₂ O ₃	1.68	2.51	1.68	6.15	0.30	4.08	-	0.85–1.23	5.94	0.18	5.92	6.87	1.84	6.81	1.98	4.32	5.67
FeO ₃	1.38	1.26	2.97	3.14	1.19	1.15	4.60	1.21–1.24	3.26	0.23	7.36	4.69	2.65	3.98	2.58	5.89	6.13
CaO	13.07	10.2	24	30.24	7.75	0.97	14.30	0.21–1.21	18.10	0.82	5	3.65	2.65	3.54	2.61	8.80	5.64
MgO	1.92	3.03	3.24	3.19	5.66	0.44	13.70	-	3.20	0.65	1.17	1.45	0.72	1.54	0.73	4.34	-
Na ₂ O	0.13	-	-	1.40	0.29	1.69	4.80	-	0.92	-	-	1.24	0.28	1.29	0.25	0.30	-
K ₂ O	38.01	42.66	31.22	12.37	3.55	1.34	2.10	-	1.85	2.03	6.22	2.59	0.32	2.17	0.31	5.70	7.51
TiO ₂	0.13	0.11	0.07	0.42	0.06	-	-	-	1.39	-	1.46	-	-	-	-	-	0.11
P ₂ O ₅	3.67	2.97	4.04	3.45	26.19	-	1.50	-	0.52	1.62	0.98	-	-	-	-	-	5.84
CL	2.01	2.26	0.07	-	-	-	-	-	0.06	-	-	-	-	-	-	-	-
LOI	25.53	18.54	23.8	-	-	6.13	6.30	3.95	15.60	17.78	9.78	4.71	10.45	4.05	9.21	-	11.20

5. The Effect of Lignocellulosic Biomass Ash By-Products on Fired Bricks

Clay bricks produced from adding lignocellulosic biomass ashes are examined in this section in terms of their physical, mechanical, and thermal properties. Only the percentage that gives a good compromise between the different physical, mechanical and thermal properties of each product has been taken into account.

5.1. Effect on Physical Properties

5.1.1. Linear Shrinkage

Linear shrinkage is a key parameter for fired bricks. It describes the contraction or expansion behavior of bricks during heat treatment. It can be determined by measuring the length of the sample either before and after drying or before and after firing. In addition, it can be evaluated over the whole process using a caliper.

$$\text{Linear Firing Shrinkage(\%)} = \frac{L_{\text{dried}} - L_{\text{fired}}}{L_{\text{dried}}}, \quad (1)$$

For the bricks produced using ashes, several factors could affect the linear shrinkage after firing either by contraction or by expansion. In general, the contraction phenomenon is related to the quantity of glassy phase formed during firing. As a result of a glassy phase being filled into the continuous voids and holes in the porous structure of ceramic material, a densification phenomenon takes place, resulting in the contraction of the elaborated product. The flux of materials such as K_2O and the auxiliary flux materials such as CaO , MgO , and Na_2O , contained in the lignocellulosic biomass ashes, could play an important role during the glassy phase. Indeed, these oxides play fluxing or melting promoters' roles allowing the cooking temperatures to decrease and to generate a greater quantity of liquid phase which contributes to the formation of the glassy phase. However, the expansion could be attributed to the dilation of pores due to the gases emitted during the firing process (mainly CO_2 and O_2). In the studies [60–62], It was observed that the addition of olive bottom ash (OPBA), olive stone ash (OSA), olive oil wastewater ash (OMW), and olive pomace fly ash (CVLL) to fired clay bricks causes an expansion condition because there is a greater effect of gaseous materials in olive waste than flow materials on the brick. In fact, as shown in Table 3, these wastes are rich in significant amounts of gaseous metals and, more specifically, significant amounts of carbonates, which decompose and release CO_2 in the pores where the phenomenon of expansion was observed for all the bricks made from the ashes of waste olives [60]. For palm oil fuel ash (POFA), Ref. [70] observed shrinkage of different samples when the POFA content increased. This shrinkage varied from 0.3% for pure clay to 0.6% and 0.9% when 1% and 10% of POFA were used, respectively. Nevertheless, the linear shrinkage increases progressively with increasing percentages of ash addition for all used ashes.

It is to be highlighted that shrinkage depends strongly on the nature and chemical composition of the biomass ash used and mainly on the content of melting oxides and continuous gaseous components in the ash, so that no real comparison could be made between the different types of ash (Figure 2). However, it is recommended that the curing shrinkage should be between 2.5% and 4% to maintain good mechanical performance [71]. Indeed, excessive shrinkage causes stresses and breaks of the prepared material as is exhibited in Figure 2 despite that all samples were manufactured with different types of lignocellulosic ashes respecting the standard conditions.

5.1.2. Loss of Ignition

Loss of ignition is the loss of mass as a result of the bricks firing. It was determined by measuring the loss of mass of the sample between the drying and firing stages. It is expressed in percentages and calculated according to the following formula:

$$\text{Loss of ignition (\%)} = \frac{m_{\text{dried}} - m_{\text{fired}}}{m_{\text{dried}}}, \quad (2)$$

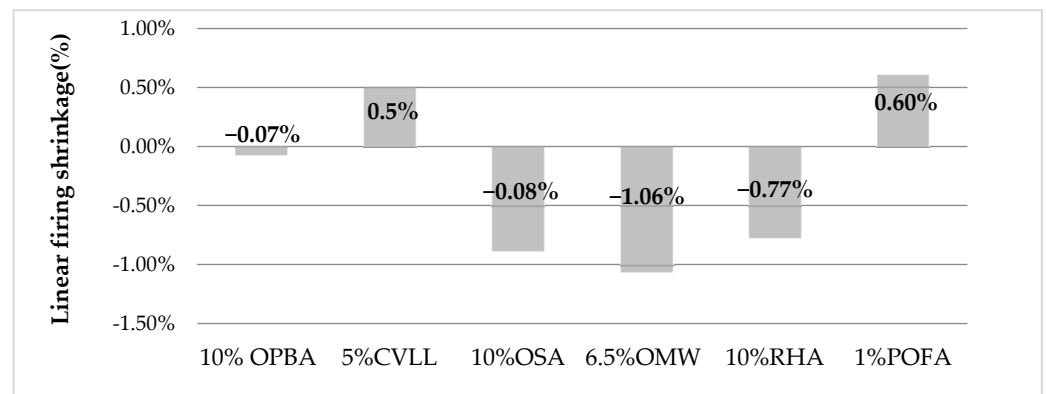


Figure 2. Linear firing shrinkage for samples made with lignocellulosic biomass ash: olive bottom ash (OPBA) [60], combustion of olive pomace (CVLL) [61], olive stone ash (OSA) [62], oil mill wastewater (OMW) [63], Rice Husk ash (RHA) [63], palm oil fuel ash (POFA) [70].

Bricks made of lignocellulosic ash lose weight due to the decomposition of organic matter and calcium carbonates, as well as the dehydration of water in the clay. When olive bottom ash (OPBA) was added in amounts ranging from 10% to 50%, the loss on ignition rose from 11.25% to 20.6%, as opposed to 10.99% for the control brick [60]. For the bricks elaborated by the waste of olive pomace fly ash (CVLL) by [61], the loss on ignition went from 12.89% for the control brick to 21.99% for 25% of CVLL addition. For [62], the addition of 30% of olive stone ash (OSA) resulted in a loss of ignition of 21.9% because the ash contained a significant amount of unburned material. Additionally, Ref. [63] demonstrated that as the cooking temperature rises, the loss of ignition of rice husk ash waste (RHA) increases even more. Additionally, in all of the studies that were conducted, it was discovered that the loss on ignition of lignocellulosic ash bricks increases with increasing the percentage of added ash in the mixture. This finding suggested that, if the sintering process is not carried out at sufficient heating rates, using these ashes to create fired bricks could result in a significant volume reduction with deformation or breakage of the bricks. Thus, to maintain good performance, the loss on ignition should remain below 15% [71].

Figure 3 illustrates that all ignition loss values are less than 15%. Due to the differences in the amounts of water used in shaping, the variations in calcium carbonate content, and the presence of organic imbricates in the ashes (Table 2), it is not possible to compare the loss of ignition between these various ashes.

5.1.3. Water Absorption

The compressive strength and durability of the elaborated samples are greatly impacted by water absorption, making it a crucial bricks parameter. For lignocellulosic biomass ash bricks, the water absorption depends mainly on the percentages of the added ashes. All of the aforementioned research projects showed that water absorption rises as the proportion of biomass ashes used in the clay matrix increases. The hydrophilic nature of biomass ash accounts for the increased ability of the hardened matrix to absorb water [70]. Additionally, the addition of the continuous organic matter in the lignocellulosic ash acts as a porogen material, leaving pores and voids during the curing process so that the open porosity increases the water absorption. The olive bottom ash (OPBA) used by [60] is a water-absorbing material. By adding this type of ash to the clay matrix, water absorption capacity is increased significantly. The increase becomes even more important when adding more than 20% OPBA. Results show that water absorption varies from 16.3% for the control brick and reaches up to 31% for 50% by weight of OPBA. This increase in water absorption could be attributed to a higher open porosity in the matrix structure. The addition of OPBA produced numerous open porosities, as well as large macropores. As a result, the open porosity of the OPBA bricks increased from 5.6% for the pure clay to 9.5% for the brick

produced by 50% by weight of OPBA. However, the closed porosity remained constant regardless of the reinforcement rate. By adding 10 wt.% OPBA, the microstructure seemed to be similar to pure clay with fewer pores, which resulted in a small increase in water absorption. According to the results of testing the olive pomace (CVLL) [61] addition to the clay bricks, higher water absorption was observed by adding 10% or 15% by weight of CVLL. In brick firing, the degradation of organic matter contained in the lignocellulosic ash is believed to be the main cause of this increase in porosity. The addition of olive stone ash (OSA) also increases water absorption [62]. Water absorption for clay bricks was 16.9%, but with 10% and 30% by weight of OSA, it increased to 19.9% and 23.1%, respectively. Regarding rice husk ash (RHA), it was found by [31,32] that adding 2% to 4% of this ash to the clay matrix reduces water absorption when compared to clay bricks and produces a good stable, and less porous structure. This character could be mainly attributed to the presence of amorphous silicon oxide which is present in large quantities in the chemical composition of these ashes (Table 2). However, percentages greater than 4% may result in a possible reduction in the surface's adhesion to the clay matrix, which is explained by an increase in RHA's granulometry as a result of which the clay matrix's porosity increases, increasing water absorption. According to [65], the water absorption produced by the addition of 30% plank wood ash (WA) was less interesting (21.9%) than the water absorption produced by the addition of 30% rice husk ash (RHA), which was 32.9%. This discrepancy results from WA ash having a higher proportion of melting oxides than other types of ash. Oxides encourage the formation of the liquid phase, which tends to approach the finer pores of the clay matrix and reduce open porosity. However, as the percentage of WA rises, this mechanism loses its effectiveness. As the percentage of WA ash increases, more open porosity, more water absorption, and less density are produced. In the same context, Ref. [67] observed a lower water absorption for bricks incorporating rice husk (RHA) compared to bricks with sugar bagasse ash (SBA). They cited SBA's porous structure as a potential contributing factor to this behavior. According to (ASTM CC67-07a, 2003) [72], the different weathering conditions can be used to categorize the water absorption of bricks. For bricks with severe weathering resistance, the water absorption should not be higher than 17%. For bricks, with moderate weathering resistance the water absorption should not be higher than 22% and no limit is set for bricks with negligible weathering.

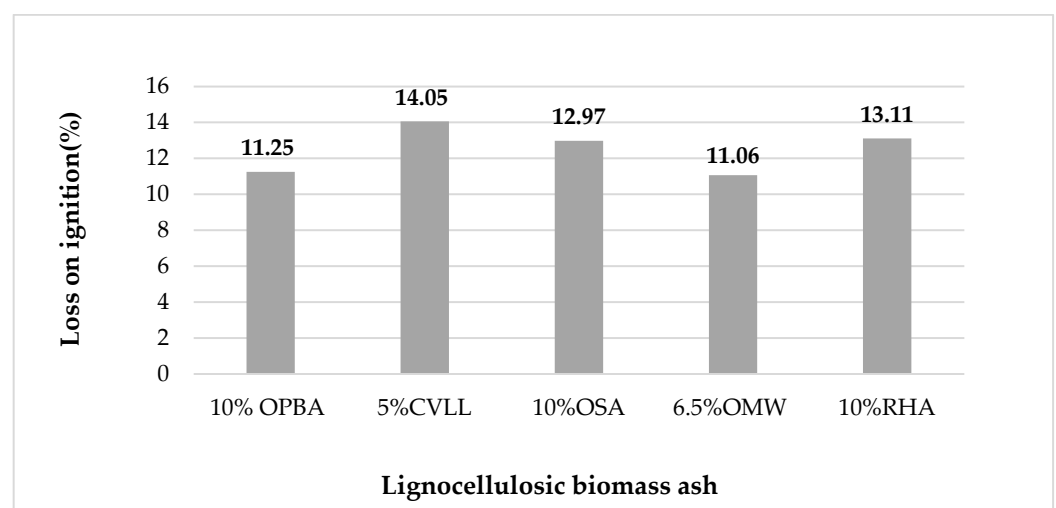


Figure 3. Loss of ignition for samples made with lignocellulosic biomass ash: olive Bottom ash (OPBA) [60], combustion of olive pomace (CVLL) [61], olive stone ash (OSA) [62], oil mill wastewater (OMW) [63], Rice Husk ash (RHA) [63].

Except for the addition of sugarcane bagasse ash (SBA) [66], which has a high-water absorption rate (>22%), all fired bricks constructed from lignocellulosic biomass ashes exhibited severe to moderate resistance to weathering (Figure 4).

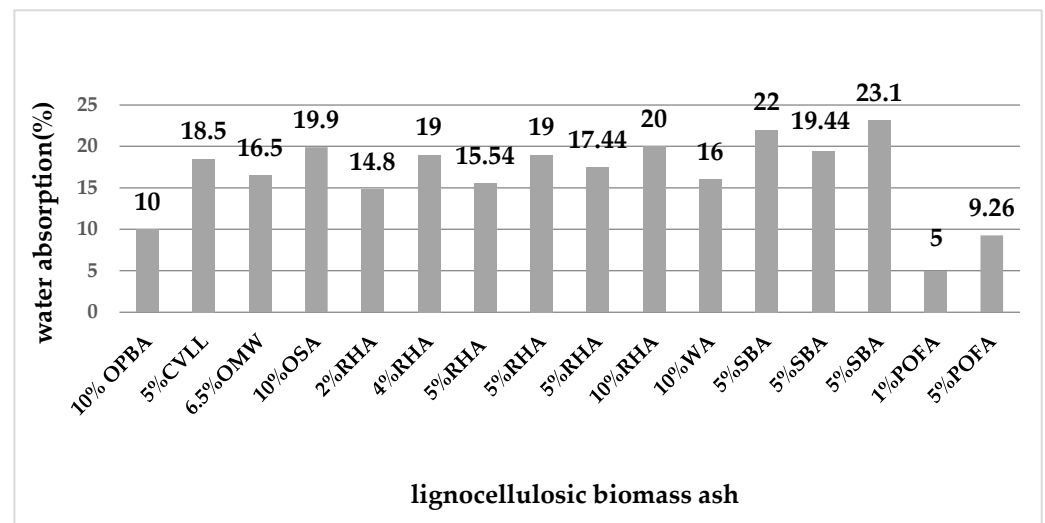


Figure 4. Water absorption for samples made with lignocellulosic biomass ash: 10% OPBA [60], 5% CVLL [61], 6.5% OMW [63], 10% OSA [62], 2% RHA [32], 4% RHA [31], 5% RHA [64], 5% RHA [67], 5% RHA [68], 10% RHA [65], 10% WA [65], 5% SBA [67], 5% SBA [68], 5% SBA [66], 1% OPFA [70], 5% OPFA [69].

5.1.4. Density

Density is defined as the ratio of the weight of the dry brick to the volume, which quantifies the amount of material present in the volume. According to this explanation, it would seem that the brick's mechanical properties and durability would be more interesting the higher this value is, while the brick's thermal properties as an insulating material would be less interesting. Bulk density averages out to 1200 to 1900 kg/m³. All of the studies showed that adding lignocellulosic biomass ash to the clay matrix decreased the bricks' bulk density, which is strongly advised for future bricks (Figure 5). Since the bricks of today are too heavy and tightly packed, research is being carried out to create new products that are lightweight and manageable during construction. Using waste lignocellulosic ash as a porogen agent and adding it to the mixture is an intriguing way to achieve the desired effect.

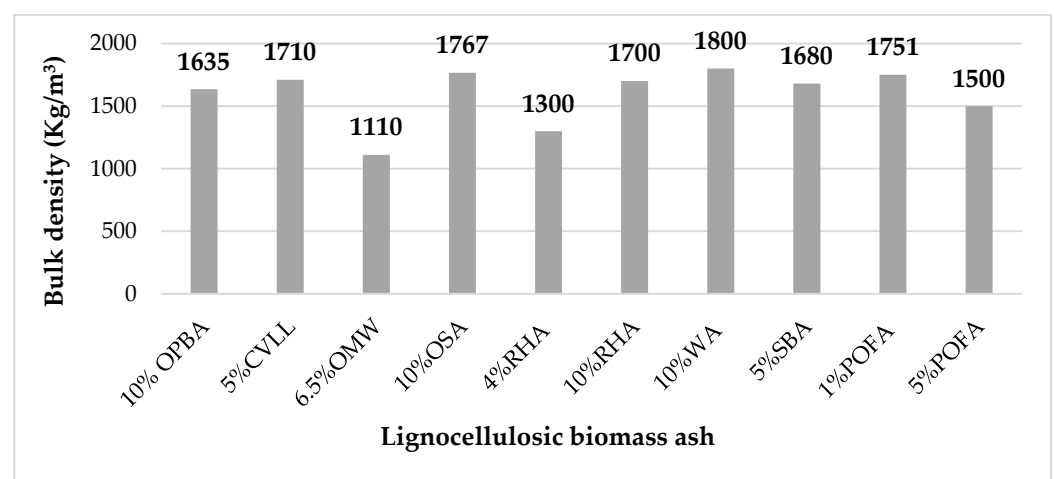


Figure 5. Bulk density for samples made with lignocellulosic biomass ash: 10% OPBA [60], 5% CVLL [61], 6.5% OMW [63], 10% OSA [62], 4% RHA [31], 10% RHA [65], 10% WA [65], 5% SBA [66], 1% OPFA [70], 5% OPFA [69].

The density of bricks made from lignocellulosic biomass ashes ranges from 1110 to 18,000 kg/m³, as shown in Figure 4. The amount of ashes used in the matrix plays a major

role in this difference. It has been established in all studies that brick density decreases as the reinforcement rate increases. However, as depicted in the diagram of Figure 5, the highest density was achieved by adding 10% of wood stone ash (WA) [65], whereas the lowest density was achieved by adding 6.5% of olive mill wastewater (OMW) [63]. The chemical composition as well as the type of combustion of these ashes largely affect the density of the final products. The incomplete combustion of lignocellulosic waste generates an increase in the content of organic matter as well as of unburned carbon in the chemical composition of the produced ashes. When these components are present during the firing of the fired bricks, they react as porogen agents and leave pores behind that reduce the density of the finished goods. The percentages of organic matter and unburned carbon in the WA ashes were 10.4% and 11.45%, whereas they were 11.61% and 46% in the OMW ashes, explaining the variations in the densities measured.

5.2. Effect on Thermal Properties

The improvement of insulation properties through the reduction of thermal conductivity of building materials is crucial for the development of energy-efficient buildings. Therefore, adding lignocellulosic biomass ash to clay bricks, which produce porosity during firing, may be a promising way to create thermally insulating bricks. Ref. [60] showed that the thermal conductivity decreases steadily as olive bottom ash (OPBA) residue is added up to 20% by weight. When 20 wt.% OPBA was added to the bricks, the values decreased to $0.82 \text{ W m}^{-1} \text{ K}^{-1}$ from $0.99 \text{ W m}^{-1} \text{ K}^{-1}$ for bricks without additives. Therefore, the addition of 10 wt.% of olive bottom ash (OPBA) resulted in a 14.1% improvement in the thermal conductivity of the bricks with a thermal conductivity value of $0.85 \text{ W m}^{-1} \text{ K}^{-1}$ (Figure 6). In addition, a similar trend was observed by [68]. They noticed that as the amount of sugar bagasse ash (SBA) and rice husk ash (RHA) in fired clay bricks increases, the thermal conductivity values decrease. They obtained a thermal conductivity at the vicinity of $0.52 \text{ W m}^{-1} \text{ K}^{-1}$. Moreover, a low thermal conductivity was observed with the incorporation of 6.5% of oil mill wastewater (OMW) [63], and an improvement of almost 65% was observed. In all the studies performed, the thermal properties of the bricks were limited to the calculation of thermal conductivity, except de Silva et al. [31] for which the measurement of acoustic insulation was performed.

Many factors such as mineralogical composition, type of porosity (open or closed porosity), pore size, measurement method, and density are important factors that govern thermal conductivity. The thermal conductivity of bricks made with lignocellulosic biomass ash is significantly influenced by the brick density. More thermally insulating bricks are produced when the density is lower. When comparing Figures 4 and 5, it can be seen that adding 6.5% of olive mill waste (OMW) produced the lowest density (1110 kg/m^3) and lowest thermal conductivity ($0.143 \text{ W m}^{-1} \text{ k}^{-1}$), whereas adding 10% of wood stone ash (WA) produced the highest density (1800 kg/m^3) and highest thermal conductivity value ($1 \text{ W m}^{-1} \text{ k}^{-1}$). This might be accounted for by the fact that a low density permits a higher percentage of air volume to fill the voids. As a result, by serving as effective insulator, the greater volume of air in the pores lowers the solid matrix's overall thermal conductivity.

5.3. What Effect on Mechanical Properties?

The most important property of construction materials is compressive strength. The compressive strength of various prepared bricks combined with lignocellulosic ashes is displayed in Figure 6. It can be observed that a reduction of compressive strength occurred when the ash content in the clay matrix increased. This result is coherent with bulk density and water absorption behavior. Bricks with higher porosity, lower compressive strength, and higher water absorption were produced at higher doses of lignocellulosic biomass ash. Due to their irregular shape and microscopic flaws, open pores can concentrate pressure and reduce the compressive strength of bricks [73].

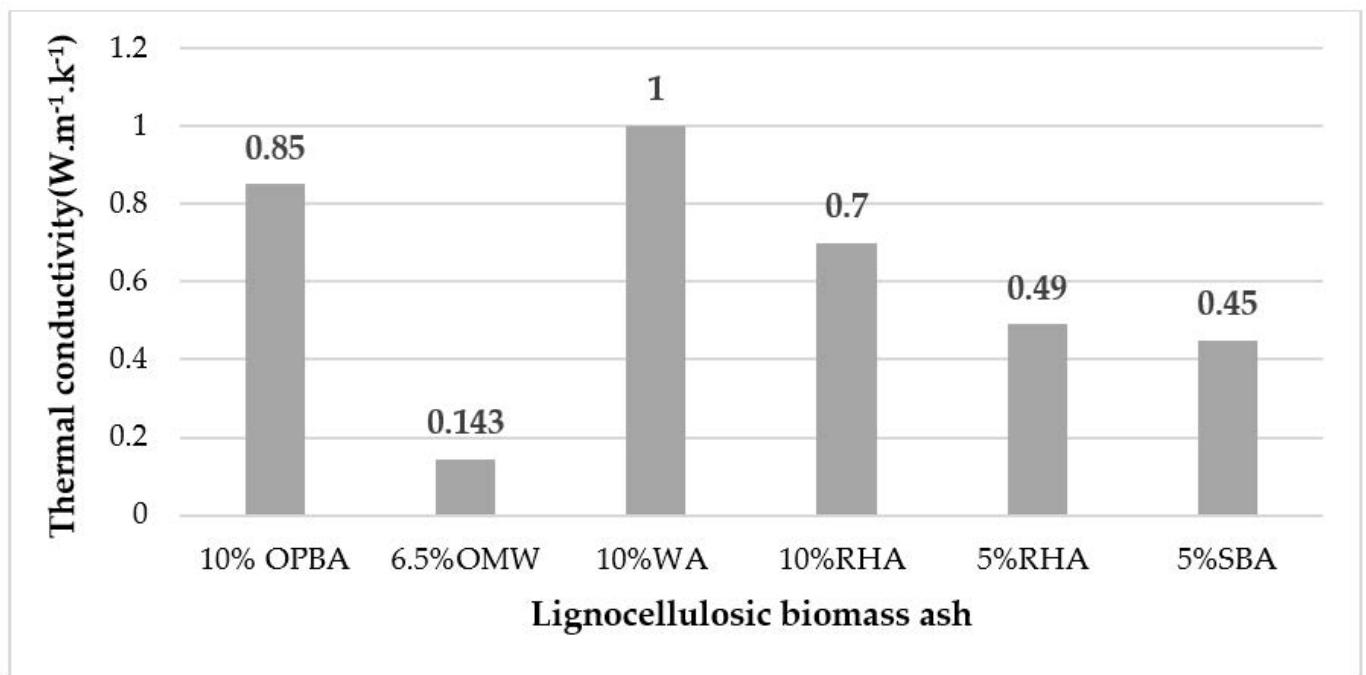


Figure 6. Thermal conductivity for samples made with lignocellulosic biomass ash: 10% OPBA [60], 6.5% OMW [63], 10% WA [65], 10% RHA [65], 5% RHA [68], 5% SBA [68].

It was noted that adding 10% and 20% by weight of olive bottom ash (OPBA), respectively, resulted in compressive strength values decreasing by 29.4% (33.9 MPa) and 71.52% (14.2 MPa) [60]. However, significant additions of 30 to 50 weight percent reduced the strength by about 78% when compared to the control bricks (47.96 MPa). For olive pomace ash (CVLL) a maximum value was reached in composite bricks by adding 5% fly ash which generates a compressive strength of 50.1 MPa [71]. Moreover, it was reported that the compressive strength results are consistent with the bulk density, bulk porosity, and water absorption data, the highest value obtained corresponds to the control bricks (53.3 MPa) [61]. Similarly, Ref. [67] noted that the compressive strength of fired clay bricks was also decreased with the addition of lignocellulosic ash. Additionally, Ref. [69] observed that as the amount of palm oil fuel ash (POFA) increased to 20%, the compressive strength decreased by 74% in comparison to the witness bricks. This fact could be related to the disintegration of organic materials during the firing process. It is to be highlighted that according to ASTM62-10 the compressive strength varies from 10 MPa for weathering bricks to 20 Mpa for high-weathering bricks [74].

As depicted in Figure 7, the values of the resistance to compression for all bricks made with olive waste ash were somewhat similar, and these bricks were distinguished by a strong resistance to weathering. On the other hand, for rice husk ash, sugar cane bagasse ash, and palm oil ash, studies showed low resistance to weathering. The compressive strength values were somewhat similar for all these ashes due to their similar chemical composition rich in fluxing oxides and silica (Table 2), except for the rice husk ash in the study by [32] which is characterized by important contents of auxiliary fluxing oxides (CaO and MgO) which increased the mechanical resistance. As previously mentioned, a variety of factors, including the source of the ashes, their chemical makeup, the type of clay used, the size of the particles, etc., determine the mechanical strength of the bricks. For instance, the ash from olive waste contains significant amounts of auxiliary melting oxides, particularly calcium carbonate (CaCO₃) and sodium oxide (Na₂O), which are crucial for the mechanical strength of brick (Table 2). In fact, during firing, CaCO₃ is what causes the release of CO₂ and CaO. When water is present, the released CaO can combine with the SiO₂ in the clay to form calcium silicate, improving the mechanical performance of fired bricks. Due to the excessive CO₂ release, which results in pore dilation, the excess of these

ashes, however, favors the appearance of cracks and expansions. Na_2O , on the other hand, acts as a melting promoter, generating a significant amount of liquid phase that contributes to the formation of the glassy phase and the densification of the fired products while filling the pores and voids.

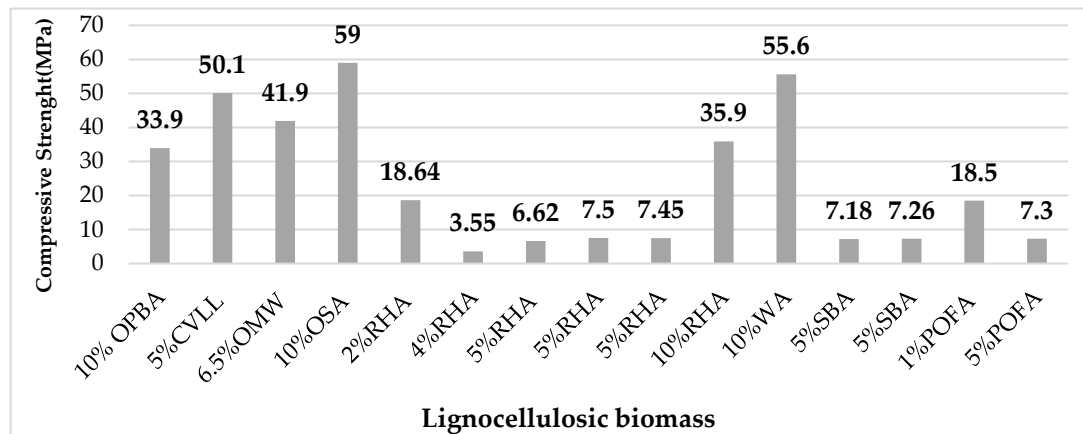


Figure 7. Compressive strength for samples made with lignocellulosic biomass ash: 10% OPBA [60], 5% CVLL [61], 6.5% OMW [63], 10% OSA [62], 2% RHA [32], 4% RHA [31], 5% RHA [64], 5% RHA [67], 5% RHA [68], 10% RHA [65], 10% WA [65], 5% SBA [67], 5% SBA [68], 1% OPFA [70], 5% OPFA [69].

Apart from the chemical and mineral compositions of soils and lignocellulosic ashes, a number of factors can influence the mechanical properties of the elaborated bricks, among which we can mention: molding technique, particle size, and during and firing process. For the molding technique, the forming pressure affects the green density of the samples, which influences the brick properties. It was observed that the higher the value of the forming pressure, the more improvement of the mechanical properties. However, this behavior ceases up to a certain limit of pressure beyond which no significant change could be observed. According to Table 2, the majority of studies do not provide pressure values [75]. Improved mechanical properties are also significantly influenced by the morphology and particle size of bricks. While finer particles create denser composites that offer better sintering and greater strength, larger particles restrict compaction within the bricks, leading to the formation of significant amounts of voids that degrade mechanical strength. Despite this characteristic, the majority of studies do not consider this influencing factor (Table 1). Using two studies by [60] and one by [61] as examples, the researchers used the same quantity, origin, and source of clay as well as the same source of ash. Figure 7 shows that the fly ash used to make fired bricks had greater mechanical strength than the bottom ash from olive pomace. This result could be explained by the fact that particles of fly ash were fine spherical powder assimilated to Additionally, the heating rate during the firing process has a significant impact on the compressive strength of fired bricks. A relatively high heating rate ($>10\text{ }^\circ\text{C}/\text{min}$) tends to form a heterogeneous microstructure, which has a severely detrimental impact on the brick's strength [76].

5.4. Currently Used Furnaces for Fired Bricks: Advantages and Drawbacks

It is suggested that the number of lignocellulosic ashes added to the clay matrix must not exceed 10% in order to achieve a compromise between mechanical and thermal performances. Furthermore, the use of wastewater from oil mills rich in melting oxides and silica with a mixture of lignocellulosic biomass ashes and small amounts of agro-food waste from the agri-food industry, such as the residues of olive by-products could be an interesting investigation. This type of substrate when added to the clay matrix, could improve the sintering and the porosity which affects positively the thermal properties of the bricks without degrading too much the mechanical properties. Despite the numerous

benefits of using lignocellulosic biomass ash in the production of clay bricks, the firing process in conventional kilns continues to be energy-intensive and polluting.

6. Unfired Clay Bricks Reinforced by Lignocellulosic Biomass Ash

Unfired clay brick construction has several benefits, such as the accessibility of raw materials, fire resistance, favorable climatic performance, low cost, and low energy consumption. These benefits combine to make these blocks a desirable building material. However, the low durability, low tensile strength, low impact resistance, low abrasion resistance, and high-water absorption capacity of these bricks are what restrict their use. Many materials, such as cement, lime, fly ash, gypsum, and bitumen, were used for soil stabilization to overcome these difficulties. In the majority of studies, cement and lime are the main components used to create stabilized soil blocks. Currently, the use of cement and lime can be offset by combining them with other wastes, such as lignocellulosic biomass ash, which is the main component of stabilized earth blocks (Table 3). Ref. [77] investigated the mechanical properties of raw earth blocks stabilized with 4% and 10% ordinary Portland cement (OPC) and altered with 4%, 6%, and 8% sugarcane bagasse ash (SBA). Except for the addition of 4% SBA, where a slight loss of strength of 3.86% could be felt, earth blocks stabilized with 4% cement show a steady increase in strength with increasing SBA content. However, a gain of 2.32% and 13.9% was observed when adding 6% and 8% SBA (2.56 MPa and 2.95 MPa, compared to 2.59 MPa of the control block). For 10% cement stabilized blocks, incremental additions of 4%, 6%, and 8% SBA led to steady increases in compressive strength of 1.48%, 2.77%, and 7.93%. Adding SBA at a lower cement content led to a greater increase in compressive strength. Adding 8% SBA was the only way to achieve a compressive strength that complied with standards for the earth block stabilized with 4% cement. However, the compressive strength was attained for blocks stabilized with 10% cement without the addition of SBA. For water absorption, the addition of SBA to cement yielded a slight increase in the water absorption of the blocks. This water absorption was higher at a cement content of 10% than at a cement content of 4% which proves that the addition of SBA is more effective at lower cement contents. In a related study, Ref. [78] prepared two stabilized earth blocks with cement contents of 6% and 12% and added sugar bagasse ash (SBA) at contents of 0%, 2%, 4%, and 8%, respectively. The choice of cement content is due to the granulometry of the soil used, thus making their blocks fall into the category of cement-bound materials. The amount of cement needed to stabilize the raw earth bricks depends significantly on the type of earth that will be used to create the specimens. While a minimum of 6% cement could be used if the earth had a significant amount of clay in it, 4% cement can produce high-performance blocks if the soil is too sandy. Strengthening to 8% SBA resulted in a maximum increase in block strength of up to 54% for bricks stabilized with 6% cement. Despite the increase in strength due to the progressive addition of SBA with 6% cement, the average values of this strength did not exceed 1.54 MPa at 28 days. For the blocks stabilized at 12% cement, an average value of the compressive strength at 28 days was 3.13 Mpa at 0% SBA. The addition of SBA results in a reduction in the mechanical strength, but this reduction is less significant for the addition of 8% SBA, or a value of 2.89 MPa was measured. Ref. [79] investigated the stabilization of residual soils using 4% to 8% cement and rice husk ash (RHA) at percentages ranging from 5% to 25% by step of 5%. When comparing the compressive strength results of stabilization of earthen blocks by adding (RHA) to cement compared to cement alone, it was seen an increase in mechanical strength. The use of 4% cement alone for stabilizing the earthen blocks leads to a compressive strength of 0.882 MPa. However, the addition of 5%, 10%, 15%, 20% and 25% RHA generates a compressive strength of 1.654, 3.154, 3.309, 3.011 and 1.187 MPa, respectively. As a result, the formula with 15% to 20% of RHA increases the compressive strength significantly when compared to the cement alone, while above this value range, the resistance significantly decreases.

In the same way, Ref. [80] considered a combination of lime, cement, and SBA for stabilizing the compacted soil blocks. The mixture was designated as NA, CAL, CEM,

and CALBA, which refer to the compacted soil blocks, respectively, when it contained no additives, 10% lime, 10% cement, and a combination of 10% lime and 10% sugarcane bagasse ash (SBA). Results of compressive strength tests show that adding cement and lime to the clay matrix increased the clay matrix's compressive strength. The cement mixture (CEM) showed a compressive strength of 12.7 MPa, while the lime mixture (CAL) showed a strength of 5.55 MPa. When SBA is added to lime in an equal amount (CALBA), the compressive strength increases by almost 40% and is measured at 7.7 MPa. This growth of compressive strength could be attributed to the progressive densification of the clay matrix and more precisely to hydration and pozzolanic reactions. For his part, Ref. [81] calculated the amount of lime required to stabilize the soil using the pH method developed by [82]. The optimal initial lime was found to be 6%. Moreover, he concluded that the stabilization could be achieved with three different sugarcane bagasse ash (BA) contents; 4%, 6%, and 8%, respectively. For this, a linear relationship between compressive strength and the percentage of BA was found, which was supported by a fit with a regression factor $R^2 = 0.94$. By adding 8% BA, this strength increased from 1.687 MPa to 1.87 MPa, or by almost 11%. Ref. [82] investigated the possibility of using lime and rice husk (RHA) for the development of new unfired brick or compressed stabilized earth (CSE) forms. To reduce the effect of shrinkage, clay as the matrix material of the brick was mixed with sand. A standard Proctor compaction test of soil and sand mixture was performed to obtain an optimal value of the sand used according to their compaction characteristics. Thus, a maximum dry density value among the test mixture was obtained for a 70% soil–30% sand mixture. The amount of lime required for stabilization (LRS) was determined according to the method developed by [82]. Accordingly, 5% lime was found to be the optimum amount of lime to stabilize the tested soil-sand mixtures. The ratio of lime and RHA was selected as 1:1 (5%:5%), 1:2 (5%:10%), 1:3 (5%:15%), 2:1 (10%:5%). It was observed that the addition of lime and RHA increases the compressive strength. This strength reaches a maximum value of 15.5 Mpa and 16.1 Mpa with a lime/RHA ratio of 1:1 for the treated clay sample and the treated clay/sand mixture sample, respectively. After this limit, the compressive strength decreases. The reason for the improved performance of clay mixed with sand is that the finer particles of clay will fill the voids of the coarser particles of sand. This would result in denser, stronger, waterproof soil mixtures which result in a decrease in water absorption. The BIS specification in IS 1725 (BIS 1982) provides for two classes of stabilized blocks, namely class 20 with a minimum allowable strength of 1.96 MPa and class 30 with a minimum allowable strength of 2.94 MPa [83]. The ideal mechanical strength for each of the aforementioned studies is depicted in Figure 8.

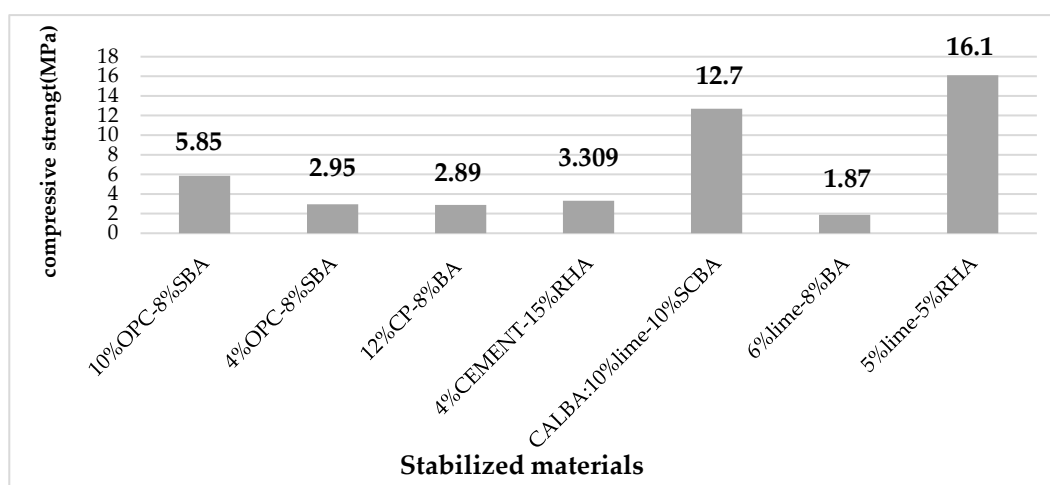


Figure 8. Compressive strength for unfired clay bricks stabilized with lignocellulosic biomass ash and lime/Cement: 10% OPC–8% SBA [77], 4% OPC–8% SBA [77], 12% CP–8% BA [78], 4% Cement–15% RHA [79], 10% lime–10% SCBA [80], 6% lime–8% BA [81], 5% lime–5% RHA [84].

The compressive strengths of all unfired clay bricks stabilized by the addition of cement/lime with lignocellulosic ash percentages, as shown in Figure 8, are higher than those of class 30, except for the study [81], where the strength corresponding to 6% lime stabilized mud block was insufficient to meet the minimum requirements of a class 20 block. According to the relationship equation developed from the fit between compressive strength and bagasse ash (BA) content, a minimum BA content of 9.42% was discovered to be necessary to achieve a strength of 1.96 MPa for Class 20 blocks [81]. In comparison to cement, this diagram demonstrates that lime is the more effective stabilizing agent. Indeed, the lignocellulosic biomass ashes are pozzolans rich in siliceous and silicoaluminous materials by themselves in powder form and in the presence of water. The silica and alumina of the pozzolans react with the calcium hydroxide continuously in the lime. Numerous hydrated calcium silicates (C-S-H), hydrated calcium aluminates, and hydrated calcium silico-aluminates (C-A-S-H) are precipitated as a result of this pozzolanic reaction. These gels formed could fulfill the voids, and give a much more impermeable structure, much denser which also translates into a gain of appreciable strength. However, several parameters could affect the mechanical properties of the pozzolanic reaction. The size of the particles is a crucial parameter. Indeed, the mixture of lime with pozzolans allows the porosity lowering of the open pores when the particles of lignocellulosic ash used are sufficiently fine. Whereas a lack of fine particles could limit both: the pozzolanic reaction and the compressive strength of the stabilized earth bricks [85]. Additionally, the presence of water increases the amount of gel that forms, favoring particle binding and increasing compressive strength [86].

The presence of carbon in the mineralogical composition may be another cause of the low pozzolanic activity in the binder-ash mixture [87]. Indeed, as a tetravalent element, carbon can both give or receive electrons. It will tend to give electrons when bonding with elements with more than four valence electrons. When there is an excess of carbon, the tendency is to form planar structures because of the ease of double bonds. The interplanar distance is therefore important and can induce water absorption decreasing the amount of water available for the hydration process of the pozzolanic reaction. This might subsequently prevent the synthesis of gels such as C-S-H and C-A-H. The color of lignocellulosic biomass ashes gives us an idea of the presence or not of carbon in its composition.

Table 3. Studies on the manufacturing of unfired bricks using waste lignocellulosic biomass ash.

Article	Stabilized Materials	Clay	Reinforcement Rate	Moisture of Molding	Forming Technique	Curing Conditions	Characteristics of Bricks
[77]	Cement (OPC)-sugarcane Bagasse. Ash (SBA) 300 μm	Solid	4–10% OPC 4–6–8% SBA	12%	Compression	Moisture cured for a period of 28 days by sprinkling water and converting it with plastic gunny bags	Water absorption Compressive strength
[78]	Limestone Portland Cement-sugarcane ash bagasse (SBA)	Sandy earth from the region of Portugal/Kaolin (1–7)	6% and 12% of cement 0%, 2%, 4% and 8% of SBA	The amount of water is adjusted for each series and it increased as the amount of SBA increased	Manual press	Room with controlled ventilation and no direct sun-light	Water absorption Compressive strength
[81]	Hydrated lime-Bagasse-ash (BA)	Virgin soil	6% of lime with 0, 4, 6 and 8% of BA	12%	The soil blocks were cast to a fixed density of 18.5 kN/m ³	Sprinkle cured for a period of 28 days and covered with plastic gunny bags	Compressive strength Water absorption
[83]	Hydrated lime (RHA)	Clay soils 70–30% sand	Lime-RHA 1:1 (5–5%), 1:2 (5–10%), 1:3 (5–15%), 2:1 (10–5%), and 3:1 (15–5%)	19%	Hand-operated compression machine of 15 MPa	Dried at humidity and room temperature (30 °C) for 28 days by covering them with a plastic sheet	Water absorption Compressive strength
[79]	Rice Husk in the incinerator ordinary Portland cement	Residual granite soil	4% to 8% of cement and 0% to 25% rice husk ash	-	Compacted with mechanical rammer of 2.5 kg	Cured in plastic bag for 7 days after 7 days of moist-curing. The specimen was then soaked for 7 days in water and the other specimen continued to cure until its old was 14 days	Compressive strength
[88]	NaOH 8M, KOH 8 M, Na ₂ SiO ₃ 8 M and KOH-K ₂ SiO ₃ 8 M	Calcined clays (CC (150 μm) Olive pomace fly ash (OPFA 150 μm)	70% CC–30% OPFA	Liquid/solid ratio = 0.4	Precursors cures at 60 °C and 98% RH for 24 h. Then specimens unmolded and stored under ambient conditions (21 \pm 2 °C and 58 \pm 2%RH) for 28 days	Bulk density compressive strength Thermal conductivity Loss of ignition	
[89]	KOH from the calcination of olive biomass ash (OBFA)	Ceramic industry (chamotte) residue 250 μm	10%, 20%, 30% and 40% of OBFA	15% and 20%	Compaction pressure of 50 MPa	In an oven at 85 \pm 5 °C temperature for 24 h	Weight loss Water absorption Boiling-water absorption Linear shrinkage Bulk density Open porosity Compressive strength

Table 3. Cont.

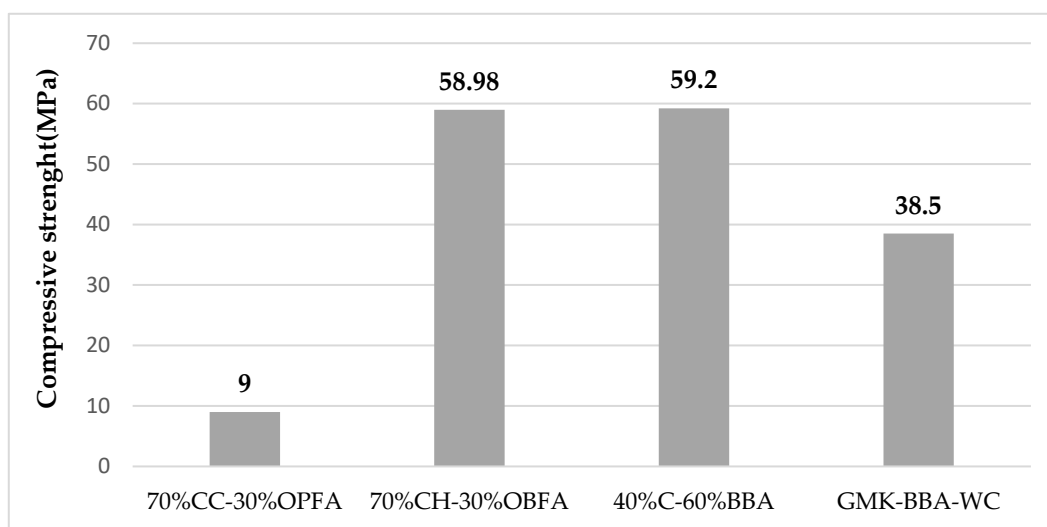
Article	Stabilized Materials	Clay	Reinforcement Rate	Moisture of Molding	Forming Technique	Curing Conditions	Characteristics of Bricks
[90]	KOH from the biomass bottom ash of almond husk and alpeorujo combustions (BBA 0.25 mm)	Chamotte 0.25 mm	10% to 100% of BBA with increases of 10%	20%	-	Dried at room temperature (20 ± 2 °C) for 24 h and at 90 ± 2 °C for another 24 h	Compressive strength Capillarity water absorption Cold water absorption Boiling water absorption Bulk density Open porosity
[91]	Biomass bottom ash residue (BBA) from the combustion of a mix of olive and pine pruning <150 μ m-sodium hydroxide 8 M	Metakaolin (MK) for different Spanish clays; Black clay(BC), Yellow clay (YC), White clay (WC) and red clay (RC) 150 μ m	33.3% of each ingredient	Water/binder = 0.6	-	The specimens were cured at 60 °C and in a saturated atmosphere for 24 h. After this period, the samples were removed from the mold and kept at room temperature until 28 days	Bulk density Water absorption Apparent porosity Compressive strength Conductivity

The stabilization of unfired bricks by chemical binders developed by alkaline activation to produce a polymerization reaction, responds perfectly to the recent challenges inherent to sustainable production. The most popular precursors used in the synthesis of geopolymers at the moment are natural clays that have undergone a thermal transformation to become calcined clays and lignocellulosic biomass ashes. The precursors of alkali activation of aluminosilicates during the geopolymerization reaction are sodium aluminates, alkali silicates, hydroxides, carbonates, and other additives. The mixture of silicate solution (Na_2SiO_3 or K_2SiO_3) and hydroxides (NaOH or KOH) is the most widely used activation solution [88] studied the feasibility of using olive pomace (OPFA) as an alkaline precursor in the manufacture of geopolymer bricks. Different samples were developed using alkaline activation solutions such as sodium hydroxide solution NaOH (8M) or potassium hydroxide solution KOH (8M), or a mixture of alkaline hydroxide and alkaline silicate solution ($\text{NaOH-Na}_2\text{SiO}_3$) and ($\text{KOH-K}_2\text{SiO}_3$) with 70% calcined clay (CC) and 30% OPFA as aluminosilicate sources, to support this proposal. A control brick was made using only water during molding with the 70% CC–30% OPFA mixture. The results showed that OPFA can be used as an alkaline activator, presenting mechanical properties slightly lower than those obtained when adding alkaline hydroxide activation solutions. The control geopolymers have a compressive strength of 1.3 MPa. However, the use of alkali hydroxides leads to an increase in compressive strength to 3.0 and 3.9 MPa for sodium and potassium hydroxides, respectively. These values increase to 5.3 MPa and 9.0 MPa when sodium and potassium silicate solutions are used, respectively. However, the best thermal insulation properties were obtained for the control geopolymers. In fact, the use of activation solutions favors the densification of the unfired bricks, which increases the thermal conductivity values. Moreover, Carrillo-Beltran et al. [89] discussed the viability of producing a geopolymer from chamotte, a byproduct of the brick industry, and olive biomass fly ash (OBFA), which is produced by calcining olive pomace (OP) and dry olive cake (DO). These ashes are composed mainly of potassium about 53% (Table 4), crystalline and amorphous constituents, and residual unburned carbon. Calcination of these ashes was carried out to promote their decarbonization while potassium oxide (K_2O) and carbon dioxide should be released. When potassium oxide is released, it dissolves in water and produces potassium hydroxide (KOH), the alkaline activation solution that leads to the formation of geopolymerization. Different amounts of distilled water (15% and 20%) and calcined OBFA (OBFA/c) (10%, 20%, 30%, and 40%) were used to prepare a group of samples. It was determined that mechanical and physical tests produced the best results for the 20% distilled water bricks. Indeed, at low water percentages, OBFA cannot dissolve properly, however, at higher values of biomass fly ash, a better dissolution could be observed due to the polymerization reaction improvement. Additionally, samples made with 30% OBFA/c and 20% distilled water demonstrate promising physical and mechanical qualities, with a compressive strength of 58.98 Mpa. However, a decrease in compressive strength was noted when OBFA incorporation exceeded 30% by weight. In the same study, Terrones-Saeta [90] investigated the feasibility of producing geopolymer using alpeorujo (BBA) (a by-product of oil composed of solid parts of olives and vegetable fats) as an alkaline activator and biomass clinker from the combustion of almond shells and Chamotte (C) as a source of aluminosilicate. The physical and aesthetic qualities of this geopolymer are comparable to those of conventional bricks. The compressive strength reaches a maximum value of 59.2 MPa for the ideal mixture of 40% chamotte (C) and 60% BBA.

Table 4. The chemical make-up of various lignocellulosic biomass ashes used in the production of unfired clay bricks.

Oxide Cotenant	SBA [77]	SCBA [80]	RHA [83]	BA [81]	RHA [79]	BBA [90]	BBA [91]	OPFA [88]	OBFA _{JC} [89]
SiO ₂	35.17	51.66	89.08	35.17	93.15	11.21	46.10	1.86	56.35
Al ₂ O ₃	0.28	9.92	1.75	0.28	0.21	2.57	12.04	0.38	14.96
FeO ₃	5.22	2.32	0.78	5.22	0.21	1.33	4.78	0.67	6.07
CaO	2.07	2.59	1.29	2.07	0.41	11.10	19.65	5.33	9.15
MgO	0.91	1.44	0.64	0.91	0.45	4.21	3.71	0.81	3.05
Na ₂ O	0.01	-	0.85	0.01	-	0.22	0.78	0.19	0.59
K ₂ O	3.75	2.10	1.38	3.75	22.31	23.91	4.59	52.1	4.52
TiO ₂	0.02	0.74	-	-	-	0.12	0.83	0.05	0.74
P ₂ O ₅	1.05	0.90	0.62	-	-	3.58	1.12	1.62	0.15

Figure 9 presents a summary of the results of the compressive strength of various investigations of the geopolymer bricks.

**Figure 9.** Compressive strength for geopolymer bricks: 70% CC–30% OPFA [88], 70% CH–30% OBFA [89], 40% C–60% BBA [90], GMK-BBA-WC [91].

It is not possible to make a comparison in these studies. In fact, several factors, including the amount of reactive phases, the chemical makeup and types of aluminosilicate sources, the nature and concentration of the alkali silicate, the curing regimes, the fillers or additive content, as well as the water content, all have an impact on the formation of geopolymer bricks.

- The influence of the clay calcination temperature To increase the reactivity of the clay used in the geopolymerization reaction, the clay must be calcined, which is a crucial step in the production of geopolymer bricks. The heat treatment transforms the crystalline phases of the clay into amorphous phases more reactive which contributes to the geopolymerization reaction and determines the final properties of the geopolymers. The optimal activation temperature depends on the mineralogical composition of the material. Thermogravimetric analysis (TGA) is commonly used to determine this ideal calcination temperature. The activation temperature must be at a temperature above the dehydroxylation peak observed on the derivative of the TGA curve or the differential thermal analysis (DTA) curve showing a downward pointing peak during dehydroxylation (endothermic reaction). One must determine the temperature that indicates the end of the peak on the DTG or DTA curve associated with the loss of

hydroxyls from the used clay and before the peak showing the structural reorganization of the mineral during recrystallization. Indeed, at very high temperatures, some crystalline minerals can appear, which decreases the quantity of the amorphous phase in the precursor and thus its geopolymerization reactivity [92].

- **The influence of the activating solution** The activating solution is an essential element in the geopolymerization process. Depending on its concentration and quantity, it will offer the right mixture to start the reaction and establish the final composition of the cured material. Additionally, it serves to speed up the breakdown of the aluminosilicate source, encouraging the development of stable gels with low solubility and the creation of compact structures using these gels. The most widely used activators are hydroxides or alkali silicates, specifically sodium hydroxide (NaOH) and potassium hydroxide (KOH). Sodium hydroxide solution is known to produce a better dissolution of the amorphous phase of aluminosilicates in combination with silicate monomers. On the other hand, potassium hydroxide solution is known to increase gel growth by association with larger aluminosilicate anions. Furthermore, the various studies conducted so far have proven that the presence of an alkali silicate solution leads to better microstructure and strength properties [93]. However, the cost of using silicate solutions during the geopolymerization reaction is high, not to mention the negative effects they have on the environment. These are prepared by dissolving glass that forms at a high temperature, which results in considerable greenhouse gas emissions [94]. It would be advantageous to use NaOH or KOH in place of the silicate solution.
- **The influence of the concentration of the alkaline solution** Generally, the dissolution of aluminosilicate sources increases with the increase of the alkaline solution concentration, thus increasing the mechanical strength of the elaborated bricks. However, the compressive strength of geopolymer materials typically declines after a certain concentration. When using high concentrations, the activation of the clay requires an excess of hydroxide ions, which can be the cause of this decrease. Indeed, this excess caused the precipitation of the aluminosilicate gel at a very early stage. As a consequence, the geopolymerization was blocked, which led to geopolymers with low mechanical properties [95].
- **The influence of compaction parameter** The mechanical performance of geopolymer materials is directly influenced by the molding and compaction process. A significant improvement in compressive strength has been noted between geopolymer bricks prepared by applying compaction pressure during the molding process and those prepared without any compression during the molding process (vibration in the vibrating table or the impact table). For the study of [88], geopolymer brick molds were prepared using 70% calcined clays (CC) of Bailen and 30% by weight of olive pomace fly ash (OPFA) as a source of aluminosilicate and a solution of potassium silicate (KOH-K₂SiO₃) as an alkaline precursor, giving 60 blows in a shock table. Compressive strength of 9 MPa was observed. Nevertheless, for the study of [89], a strength of 59.2 MPa was determined for geopolymer bricks prepared entirely from chamotte, the residue of the ceramic industry as a source of aluminosilicate, and potassium oxide (KOH) contained in olive biomass fly ash as an alkaline solution, a compression of 50 MPa was used to mold specimens of an internal dimension of 6 × 3 cm. However, to achieve the 50 MPa for bricks with actual dimensions of 22 × 10.5 × 5 cm³, 1155 KN of force must be applied, which is completely unsuitable for use on an industrial scale.

7. Unfired Bricks: Advantages and Drawbacks

In comparison to fired bricks, stabilized unfired bricks permits the incorporation of higher levels of lignocellulosic biomass ash waste than fired bricks. The majority of unfired bricks are stabilized by the use of cementitious binders (lime/cement), but the high carbon footprint associated with the use of these binders is considered the main

drawback. Geopolymer bricks were recently developed in the ceramic industry and were considered viable alternatives to cement-lime stabilized bricks. The stabilization of bricks by chemical binders using alkaline activation fits perfectly in the sustainable development context where a high volume of waste such as lignocellulosic biomass ashes could be reused and valorized in the clay matrix with a carbon footprint considered as zero. Indeed, as geopolymers have molecular structures close to zeolite, they can also immobilize toxic wastes or heavy metals contained in these ashes, thus decreasing the cost and the problem of burying this type of material. Nevertheless, different drawbacks limit the use of these geopolymer bricks on an industrial scale. Indeed, the alkaline activation of calcined clay is the most classical way and the most used in research to obtain a geopolymer with good mechanical properties and durability. Indeed, the calcination of clay allows for to modification of its crystalline structure into an amorphous structure and then improves its reactivity in an alkaline medium. Alkaline activation of clay soils at low temperatures is being researched more and more as a method of soil stabilization. Indeed, the use of low-quality raw clays other than kaolin and without thermal pretreatments, such as Montmorillonite, Illite, etc. would be a solution to reduce greenhouse gas emissions, manufacturing costs, and excessive use of kaolin by valorizing widely available natural clays at low-cost. This substitution would allow an energetic gain by overcoming the step of thermal treatment of clay. However, the understanding of the reactivity of soils using alkali activation stabilization is complex because of the different parameters that can influence their reactivity such as the clay composition of the soil, and the particle sizes, . . . In this context, a better understanding of alkaline activation of clay minerals without prior calcination is needed. The commercialization of these geopolymer bricks may also be constrained by the high cost and negative environmental impact associated with the use of alkaline activators in various research projects. The use of mixed binders (chemical and cementitious) seems to be appropriate to overcome the cost considerations for chemical binders. However, the carbon footprint of using cementitious binders is still there [96]. As an alternative to commercial alkaline activator solutions, research is focused on the use of lignocellulosic biomass ash, primarily olive waste rich in potassium oxide. These ashes are inexpensive, environmentally friendly, and can be used to create long-lasting, unfired clay bricks with good mechanical and physical qualities. These geopolymer bricks based on lignocellulosic ashes could compete with fired bricks which are preferred by their high mechanical resistance.

8. Conclusions

This study focuses on the use of lignocellulosic ash as a partial replacement for clay in the manufacture of fired and unfired clay bricks and their industrial integration. Indeed, the use of lignocellulosic ash in the manufacture of bricks is a promising way to decrease the need for non-renewable clay, as well as to reduce the environmental risks and high costs associated with the landfilling of this ash. Several conclusions can be drawn:

- Ashes from lignocellulosic biomass are favored by high concentrations of melting oxides, which enable the reduction of the firing temperature of the fired clay bricks. However, a sizable amount of their use causes the bricks' compressive strength to decline. Considering the various results of the research made on fired clay bricks elaborated by lignocellulosic biomass ashes, and to make a compromise between thermal and mechanical properties, it is advised that the rate of reinforcement within the clay matrix be lower than 10% for the ashes-rich in melting oxides and lower than 4% for the ashes rich in amorphous silica and gaseous materials to ensure a compromise between physical, thermal, and mechanical properties. According to the papers studied, ash of rice husk, sugar cane bagasse, and palm oil contains high levels of amorphous silica (more than 50%). These materials have low compressive strengths (7.5 MPa), except in [65] where high compressive strengths (35.9 MPa) were found due to the high pressure (54.5 MPa) used in molding the brick.

- To decrease the energy required to fire bricks in conventional kilns, numerous studies have concentrated on the development of unfired bricks stabilized by lignocellulosic biomass ash and cementitious binders (lime/cement). The combination of cement and lignocellulosic ash offers a promising way to both cut down on cement usage and waste production. The addition of lignocellulosic ash at a lower cement content results in the gain of higher compressive strength. With cement percentages below 10%, this gain was no longer significant. Despite the effectiveness of adding these ashes to cement to improve mechanical properties, using lime with lignocellulosic ashes demonstrates greater efficiency than cement because of the pozzolanic reactions that occur and cause the clay matrix to gradually become denser.
- New research has been focused on the stabilization of bricks using chemical binders by alkaline activation or by a geopolymer mechanism to produce more environmentally friendly and sustainable building materials and to lessen the significant carbon footprint associated with the use of cementitious binders for the stabilization of unfired bricks. The use of lignocellulosic biomass ash rich in potassium oxide is a promising and affordable solution to reduce the high cost of chemical binders in the manufacture of geopolymer bricks. It has interesting mechanical and physical properties similar to those desired for fired bricks, which encourages brick factories that are a part of the sustainable development framework to carry out this research from the laboratory scale to the industrial scale.

9. Perspectives

For the brick industry, one of the top priorities is to be as environmentally friendly as possible. Doing so will result in less energy use and fewer greenhouse gas emissions. The use of lignocellulosic biomass ashes rich in molten oxides is undoubtedly an effective way to lower the firing temperature, but geopolymer bricks based on potassium-rich ashes are the bricks of the future. However, standards set the limits for the use of these bricks in the ceramic sector, and leaching tests to have the environmental impact of the use of lignocellulosic biomass ashes in the manufacture of geo-polymer bricks must be performed. Indeed, the standards used to characterize these types of materials are currently those used for Portland cement, which can lead to an overestimation or an underestimation of the properties of these materials. However, expert researchers in the field of construction materials, grouped within the RILEM TC AAM scientific committee, are in the process of pooling their efforts to propose standards for the formulation and characterization methods of this type of material.

Funding: This research received no external funding.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Conflicts of Interest: The authors declare no conflict of interest.

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