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# The recycling of demolition roof tile waste as a resource in the manufacturing of fired bricks: A scale-up to the industry

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#### ABSTRACT

This study illustrates the utilization of roof tile waste as a resource in the manufacturing of fired bricks. Although commonly referred to as demolition waste, it is technically classified as construction and demolition waste (C&D). This demolition waste was used as a partial replacement of two soils (alluvial and laterite soil) at three firing temperatures that were considered economical (700, 850, and 900 °C). The waste considered was obtained from roof tiles previously fired at a low temperature below 800 °C, thus containing residual carbonates and clay minerals. The increased waste input resulted in higher firing shrinkage, bulk density, and water absorption while decreasing loss on ignition. An increase in firing temperature led to higher firing shrinkage, loss on ignition, and bulk density, but lower water absorption. The bricks met both Indian and ASTM standards for 2nd and 3rd class by adding 20–35 wt% of roof tile waste and firing at 850–900 °C in laboratory and industrial settings. The minimum acceptable quality for the produced bricks was achieved with an addition of 35 wt% waste, resulting in a water absorption of approximately 19% and a compressive strength ranging from 6 to 9 MPa. The study suggests that incorporating waste from demolished roof tiles into the production of burned bricks can be advantageous. It can partially replace the need for soils, reduce natural resource usage, lower energy consumption during production, and decrease the carbon footprint.

# 1. Introduction

Brick has been the most common building and construction material since 4,5000 B.C. when the Romans first used it [1,2]. Clay has the potential to be utilized for the production of burnt bricks due to its chemical, mineralogical, mechanical, and physical properties. Clay is also a suitable material due to the qualities that make it a significant option for incorporating different sorts of waste when creating burnt bricks [3]. Research into the use of waste materials in the brick-making process is an ongoing endeavor that covers a considerable time frame [3]. Bricks can be manufactured using a variety of waste products, each of which has the potential to positively influence the mechanical and physical properties of the finished product, but can also lead to somewhat undesirable properties [3]. Construction and demolition (C&D) waste has a significant influence on the overall amount of dumped material in the world. The composition of this waste exhibits complex behavior and requires further research [4].

The term "construction and demolition waste" refers to materials and products that are damaged or useless as a result of repair, demolition, and other construction-related activities [1,5]. This waste accounted for 30% of all the waste produced in the EU and 40% of the total municipal solid waste [6,7]. The C&D activities are considered the main source of waste in 2020 [8]. It is made up of a variety of materials, mainly mineral waste (concrete, bricks, roofing tiles, ceramic tiles, etc.), and then metal, glass, wood, gypsum, paper, and cardboard, mixed and hazardous waste [9]. Recycling percentages range varies from 10 to 90% among EU member states [2,10]. The highest material proportion is concrete, which ranges from 38% in Northern Europe to 61% in Western Europe. The proportions of wood and brick in Northern Europe are about 5% and 15% respectively [11,12]. Steel fractions range from about 4 to 6%, whereas the largest percentage of plastic is found in Southern Europe (2.3%) [11]. As one of the main sources of waste, which is officially landfilled in Europe in 35%, but also ends up in wild dumps reaching a total of 54% [6,11], these materials are among the top five waste

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products by quantity, and what's even more worrying is that this is predicted to increase in tandem with economic expansion [6,7]. While recycling is of high priority, notwithstanding the significance of environmental considerations, user preferences indicate that the price/quality relationship remains the main determinant of consumer choice [13]. The construction and building sectors take a heavy toll on the environment. According to the United Nations Environment Programme, this sector is responsible for 30% of natural resource extraction, 25% of waste generation, 25% of water consumption, and 12% of land exploitation [14]. The building and construction industries are creating different levels of burden on the natural environment like releasing CO2 emissions due to the energy consumption of machines and dust particles into the air by building production industry processes, and highly utilizing natural resources by the production of construction materials such as cement, bricks, steel, aluminum, and so on [15,16]. When comparing the environmental impact of manufacturing 1 kg of Ordinary Portland Cement (OPC) and 1 kg of clay bricks, it is found that 1 kg of OPC consumes roughly 0.49 kWh of energy and emits 0.65 - 0.92kg of CO<sub>2</sub> [17]. On the other hand, the production of 1 kg of clay bricks requires 2 kWh of energy and releases 0.41 kg of CO2 per brick into the atmosphere [1]. Globally, the whole construction industry is considered responsible for about 39% of the annual CO<sub>2</sub> emissions [11]. At the meeting in Paris in 2015, the United Nations set the Sustainable Development Goals for 2030 and 2050 to reduce, recycle, and increase the resource efficiency of C&D waste [18]. The majority of waste, 83%, is produced during demolition, with 17% coming from renovation [11].

Since demolition waste is the leading source of C&D waste, its utilization is considered an emerging research area. Most of the studies are focused on concrete technology, aggregate, and partial replacement of cement and sand [19]. Meanwhile, studies on the usability of C&D waste in fired brick production are rare. Furthermore, no regulations or rules exist to utilize the waste to make fired brick. Fine-grained fractions (<0.125 mm–0.6–0.125 mm) of C&D residues obtained by milling are successfully implemented in 15 wt% at laboratory-scale tiles produced by the extrusion process [12]. In another study, a fraction of below 0.6 mm of C&D waste proved useful to be implemented in levels of 10-25% to produce hand-molded bricks [20]. Furthermore, 20 wt% of waste glass and 10 wt% waste demolished brick fractions below 105  $\mu$ m were successfully mixed into the clay to make hydraulically-pressed bricks [61].

Roof tile waste (RTW) is generated during the demolition and renovation of buildings. It is one of the C&D wastes, and several research works have been done on incorporating it into construction and building materials. In Class-F fly ash concrete, roof tile waste is employed as a coarse aggregate in the internal curing agent. As a result, the compressive strength increased and the elastic modulus of the recycled concrete decreased. Additionally, the hydration of cement paste is positively impacted [21,22]. In another study, terrazzo roof tile waste was studied as the precursor for the development of geopolymer binders [23]. The alkali activator to binder ratio, the molarity of sodium hydroxide, and the elevated curing temperature are parameters that showed a significant effect on the polymerization process, whereas the weight ratio of sodium silicate to sodium hydroxide appeared insignificant [23]. Furthermore, roof tile waste was studied as wall cladding material and showed a promising positive effect for low-income enterprises A specific blunting treatment has been found to turn the tile fragments into artificial stone which is further treated to serve as a decorative accent for non-load-bearing vertical surfaces [24]. Recycling of roof tile waste has not yet been investigated in fired brick making so this opportunity could be studied and implemented as a waste minimization option for the globe.

The investigation in this work focuses primarily on the exploitation of waste as a resource for the manufacturing of fired bricks. The major objectives that this research aims to achieve are recycling the waste from demolished buildings into sustainable resources, making efficient use of the waste in the construction and building industries, and producing

fired bricks by partially using this waste as an alternative to raw clay.

# 2. Materials and methods

# 2.1. Materials

This study combined two types of soil and demolition RTW to create a fired clay brick at three peak firing temperatures. The two soils utilized in the production of bricks were alluvial soil (AS) and laterite soil (LS). The collection and preparation of the soils were the same as in earlier studies [25]. For all the materials, a fraction below 0.6 mm was used. The used LS contained 13.08% of the clay-, 47.47% of the silt-, and 39.46% of the sand-sized fractions [26].

Alluvial soil is one of the natural resources that has the widest range of applications in the construction and building sector. The physical properties, structure, and composition of the soil make it appealing for agricultural reasons, fine ceramics, and brick manufacturing. The texture varies from sandy clay to silty clay, or even clayey in the case of river delta locations [26]. Likewise, the structure varies, with sandy soils being loose and free-draining and clayey soils being dense and impenetrable. The color of the soil ranges from yellowish to grey and greyish brown [20,25,26]. This soil is rich in humus and potash but is deficient in phosphorus and nitrogen [58]. Another feature that draws attention to the manufacturing of fired clay bricks is the glass phase of the AS [27]. The AS employed in this study contained 17.73% of the clay-, 72.22% of the silt-, and 10.05% of the sand-sized fractions [26].

LS is found in regions with high annual precipitation. It has a light texture, an open free-draining structure, a low lime concentration, and also some gravel, making it ideal for brick production. Iron ( $Fe_2O_3$ ) is a component of the chemical makeup of the soil, which is responsible for giving it a reddish color.

According to Indian standards [25,26] laterite and alluvial soil are recognized as the two most significant natural resources for the brick-making industry in India. These soils are crucial to the sector as they contain a large percentage of clay minerals and fine particles in their composition while low amounts of nitrogen, phosphorus, and organic matter are present. Both soils' chemical and mineralogical constituents are shown elsewhere [28].

The demolished RTW was collected from various locations both inside and outside of the Indian Institute of Technology Guwahati (IITG) campus. The waste was generated as the result of the process of dismantling the construction of a building or other structure, which ultimately led to the generation of the waste. The preparatory work was done in a step-by-step manner once the waste from the roof demolition had been collected. The first step in the preparation process was the separation of the unwanted materials from the demolition waste, such as bricks, concrete, plastics, and other materials. Crushing and pulverizing the material were the next two processes once this stage was finished. The crushing was done manually with a hammer, whereas the dry milling was performed using a mill with tungsten carbide balls (Herzog HSM 100 H, Germany). After milling, the fraction passing through a sieve with a suggested size of 0.6 mm, which is the same as the size of the two distinct types of soil, was accepted for further tests.

# 3. Methods

The production of fired bricks follows a technique that may be divided into two major parts for easier comprehension. Characterization of the raw materials was completed first, followed by a laboratory-scale analysis and a commercial-scale assessment of the bricks. Raw material characterization included mineralogical, chemical, and thermal examination of the soils and RTW.

PANalytical AXIOS Sequential X-ray fluorescence (XRF) Spectrometer was employed to examine the chemical composition (main and minor oxides). The calibration of the instrument is done by using certified reference materials. Loss on ignition is determined by

calcinating the material at 1000  $^{\circ}\text{C}$  for 1 h, and later on these results are used to calculate the chemical composition.

The Fourier transformer infrared spectroscopy (FTIR) spectroscopy (IRAffinity-1; M/s Shimadzu, Japan) was applied to determine the functional groups that were present in the raw material. At room temperature, measurements were taken at a resolution of 4 cm<sup>-1</sup> in the midinfrared spectral region. Up to ten scans per second were conducted at a high scan rate. The fast recovery deuterated triglycine sulfate (DTGS) detector with automated atmospheric suppression and the KBr beam splitter were used. The FTIR spectrum graph was created by using four unique modes of molecular vibration (bending, rocking, twisting, and scissoring) by which it presents the functional groups in a compound. FTIR is useful in identifying and characterizing unknown materials, detecting contaminants, finding additives, and identifying decomposition and oxidation [29].

A Rigaku Technologies, Tokyo, Japan X-ray diffraction (XRD) machine has the potential to make electron density maps with an accuracy that is on par with that of atoms by using X-rays with a wavelength of 1.54184 Å. The diffractograms were taken in the 5–90 $^{\circ}$  2-theta range with a step of 0.05 $^{\circ}$  and an exposure time of 6 s per step. The EVA v.9v0 software package and the PDF-2 crystallographic database were employed. Mineral reflections were adjusted by dividing by the corundum reflection's strength.

Differential thermal and thermogravimetric analysis (DTA/TGA) Netzsch STA 449F3A00 instrument was employed in an investigation into the weight change and thermal stability induced by a firing temperature heating rate of 10  $^{\circ}$ C/min in a static nitrogen environment up to 1000  $^{\circ}$ C. The weight of the tested samples was 15 mg.

A Zeiss Sigma 300 scanning electron microscope with energy dispersive X-ray spectroscopy (SEM-EDX) was used to analyze the microstructural morphology and elemental composition of waste from demolished roof tiles and the magnification range from 10 to 300,000 times. The sample was properly dried and gold-coated to avoid electron charging during analysis.

Finally, the Toxicity Characteristics Leaching Procedure (TCLP) technique was used in this study to determine the quantities of heavy metals present in the demolition of RTW. The Environmental Protection Agency (EPA) identifies the following metals detection procedure [30]: arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), nickel (Ni), manganese (Mn), lead (Pb), and zinc (Zn).

After the raw materials had been characterized, the waste was incorporated into the manufacturing process of fired clay bricks. Various

percentages of waste from demolished roof tiles are mixed into the two different kinds of soil (5 to 45 wt%, by an increment of 5 wt%). For this study, temperatures of 700, 850, and 900 °C were used. Brick manufacturing was carried out in two distinct ways: one for the addition of waste to LS and another for the addition of waste to AS. After the proper dry mix had been completed, the next step was to prepare the wet mix, which requires the addition of water ranging from 20 to 25 wt% to achieve the desired consistency and provide the brick with the plasticity required to be poured into the mold. The laboratory-sized  $(61 \times 29 \times 19)$ mm) and industrially-sized molds (230×110×70 mm) were used, and the samples were formed by hand. The complete procedure of producing the bricks of all sizes was the same. The top cover of the mold for industrial samples had engraving (Fig. 1). The bricks are allowed to air-dry in the sun for one day before being dried in an oven at 50 °C for 6 h and later on at 105  $\pm$  5  $^{\circ}\text{C}$  for 24 h. Sintering of the brick in the muffle furnace has been done for one hour at the required temperature, while it takes a maximum of three hours to attain the requisite temperature. For each mixing ratio, eight specimens were made, and controlled bricks were also produced utilizing alluvial and laterite soil for laboratoryscaled bricks. The manufacturing process is illustrated in Fig. 1.

The two stages of the fired brick examination were carried out. The first stage was an analysis at a laboratory-sized level, while the second phase employed the analysis of commercially scaled bricks. Compressive strength, water absorption, linear shrinkage, loss on ignition, and bulk density measurements have been conducted on laboratory-scaled burnt bricks. On the other hand, the compressive strength, water absorption, and bulk density characteristics of commercial-size brick were investigated.

The firing linear shrinkage was measured by a digital vernier caliper to provide an accurate reading of the volume change that occurs after the muffle furnace. The loss on ignition determination provided information on the amount of weight lost due to the evaporation of the organic components, carbonates, and clay minerals disintegration that occurs during the firing process. It was calculated as the difference in weight between the specimens before and after being heated in the muffle furnace. After determining the volume of samples, the bulk density was calculated.

A Universal Test Machine (UTM) was used to determine the compressive strength of laboratory-scaled bricks. The compressive test was performed by exerting a stress of 250 kN onto the sample in a continuous manner at a rate of 0.2 kN/s until it completely breaks. The UTM machine has an automated adjustment system that ensures a



Fig. 1. Manufacturing of laboratory- and industrially-sized bricks.

consistent distribution of weight over the specimens. Following the standards established by ASTM C62–13a [31] and IS, 3495–1 to 4,1992 [32], the final presented result was determined as a medium of five samples. At room temperature, three samples of the brick were soaked in water for 24 h. Afterward, the bricks were taken out, wiped down with a moist towel, and then promptly weighed after being put on the scale.

# 4. Results and discussion

In this chapter, all of the experiments on raw materials and final products of fired brick manufactured with the integration of demolished RTW are displayed with references to their respective explanations. The main stages were the characterization of the raw materials and the examination of the finished products. Since the characterization of the used AS and LS is shown in the previous literature [20,25,26,28], the emphasis in this work was on defining the important characteristics of the waste from demolished roof tiles. The second part focuses on the experimental outcomes of the products (fired bricks). Both laboratory-scale and commercial-size bricks are tested in the process.

# 4.1. Raw material experimental results

# 4.1.1. Chemical and mineralogical composition

The chemical composition of RTW and the mixed materials containing AS and LS and 5 and 45 wt% of the waste are presented in Table 1. The XRF result of the alluvial and laterite soils is presented in a previous study. AS contained 47.07, 28.64, and 5.43 wt% of SiO<sub>2</sub>,  $Al_2O_3$  and  $Fe_2O_3$ , respectively, with a loss on ignition of 9.62%. LS contained 46.07, 26.86 and 10.58 wt% of SiO<sub>2</sub>,  $Al_2O_3$  and  $Fe_2O_3$ , respectively, with the loss on ignition of 8.31% [3].

The main oxides present in the materials were  $SiO_2$  and  $Al_2O_3$ . These are the two components that are crucial for producing high-quality burnt bricks [33]. In the production of burnt bricks, one of the criteria to determine the suitability of raw material is a mass ratio of  $SiO_2$  to  $Al_2O_3$  which should fall between 0.5 and 4.5. If there is an excessive amount of  $SiO_2$ , this ratio will be larger (4.5). Nevertheless, in the case it

Table 1
Chemical composition (XRF results) of the roof tile waste and boundary mixtures.

	RTW	AS+ 5% RTW	AS+ 45% RTW	LS+ 5% RTW	LS+ 45% RTW
SiO <sub>2</sub>	59.51 ± 3.79	47.69 ± 3.17	53.91 ± 3.34	$46.74 \\ \pm 3.12$	53.46 ± 3.31
$Al_2O_3$	13.84	27.90	20.50	26.21	19.70
Fe <sub>2</sub> O <sub>3</sub>	$\pm 0.88$ 5.64	$\pm 1.80$ 5.44	$\pm 1.17$ 5.55	$\pm 1.72$ 10.33	$\pm 1.01$ 7.86
- 2 - 3	$\pm \ 0.41$	$\pm 0.39$	$\pm 0.41$	$\pm~0.68$	$\pm 0.53$
MnO	0.05	0.53	0.28	0.13	0.56
	$\pm 0.00$	± 0.00	$\pm 0.01$	± 0.00	$\pm 0.02$
MgO	$2.57 \pm 0.21$	$2.02 \pm 0.19$	$2.31 \pm 0.20$	$1.87 \pm 0.12$	$2.24 \pm 0.16$
CaO	± 0.21 7.14	± 0.19	± 0.20 4.44	± 0.12 1.69	± 0.10 4.56
	± 0.52	$\pm 0.12$	$\pm$ 0.42	$\pm 0.14$	± 0.43
Na <sub>2</sub> O	0.99	0.82	0.91	1.03	1.01
	$\pm~0.07$	$\pm 0.07$	$\pm~0.07$	$\pm~0.08$	$\pm 0.08$
K <sub>2</sub> O	$3.79 \pm 0.25$	$3.73 \pm 0.25$	$3.76 \pm 0.25$	$2.82 \pm 0.18$	$3.33 \pm 0.23$
${\rm TiO_2}$	0.55	0.66	0.60	0.50	0.53
	$\pm 0.04$	$\pm 0.04$	$\pm~0.04$	$\pm~0.03$	$\pm~0.04$
$P_2O_5$	0.07	0.22	0.14	0.26	0.16
60	± 0.01	$\pm 0.01$ $0.11$	± 0.01	$\pm 0.01$	± 0.01
$SO_3$	$0.06 \pm 0.01$	$\pm 0.01$	$0.08 \pm 0.00$	$0.18 \pm 0.01$	$\begin{array}{c} 0.12 \\ \pm 0.01 \end{array}$
Mass ratio SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub>	4.30	1.71	2.63	1.78	2.71
Loss on	5.55	9.42	7.38	8.17	6.79
ignition, 1000 °C	± 0.43	± 0.56	± 0.49	± 0.53	± 0.45

is lower (0.5), there will be a greater presence of  $Al_2O_3$  [34,35]. This ratio was found satisfying in all the mixed materials and the RTW alone (Table 1). The addition of RTW influenced an increase in the SiO2 and a decrease in Al<sub>2</sub>O<sub>3</sub> contents. The situation with Fe<sub>2</sub>O<sub>3</sub> is favorable: its content achieved a mild increase when AS was used, and a significant decrease in the case of LS. The mixtures are found as potentially useful as noncalcareous as red clays [36]. Due to the significant presence of fluxing oxides in the RTW, the sintering and melting points of the brick are lowered, which in turn speeds up the firing process. Because the sum of fluxing agents (Fe<sub>2</sub>O<sub>3</sub>, CaO, Na<sub>2</sub>O, K<sub>2</sub>O, and MgO) were all present in a quantity above 10 wt%, which was 14.49 wt% in the waste, this indicates that the inclusion of these materials will lower the needed firing temperature of the products that will fulfill the quality requirements [37,38]. The sum of fluxing oxides is increased in all the mixtures, compared to the raw clays. The loss on ignition (LOI) in the RTW was 5.55%. The greater LOI may indicate leftover organic materials present in the RTW, the presence of carbonates, and/or further dehydration of clay minerals [39]. Since the LOI values are decreased while using RTW, a lower carbon footprint of these products is expected. Considering that the local roof tiles are fired in between 700-800 °C, this result was expected but tested further by other instrumental techniques.

Fig. 2. Depicts the distinct molecular bands detected by FTIR, which were found at about 3389, 1645, 1463, 1000, 783, 562, and 460 cm $^{-1}$ . The bands that indicate the presence of hydrogen bonds can be found on the bands between 3650 to 3250 cm<sup>-1</sup>, which include amino, hydrate, and hydroxyl compounds. Due to a low quantity, the band intensity of an O-H group at 3389 cm<sup>-1</sup>, which is assigned to water adsorbed on illitemica and Ca(OH)2, was not sharp [40]. The band at a wavelength of 1645 cm<sup>-1</sup> presents a nonlinear molecule with an O-H bond deformation (bending vibration) in the illite-mica minerals [41]. The appearance of a band at 1463 cm<sup>-1</sup> is assigned to CaCO<sub>3</sub> [20]. The most prominent peak, which mostly corresponds to quartz as a dominant component in the tested material, is found at 1000 cm<sup>-1</sup>. The characteristic singlet and doublet, positioned at about 750-780 cm<sup>-1</sup>, respectively showed the presence of crystalline and amorphous forms of quartz [41]. The band at 562 cm<sup>-1</sup> corresponds to trace amounts of hematite [41]. The stretching vibration associated with the band at about 460 cm<sup>-1</sup> is a Si-O-Si stretching vibration of quartz [42].

XRD analysis was carried out to get a complementary understanding of the mineral phases and identify the crystalline structure of the raw materials. The sharpest and most prominent peaks in the XRD graph are attributed to the abundant presence of quartz (SiO<sub>2</sub>), especially in the tested waste material, (Fig. 2) This result confirms those obtained by XRF (Table 1) and FTIR Fig. 3. and is following the nature of the usual raw materials for the production of roof tiles [43]. Furthermore, some illite-mica and feldspars (albite and orthoclase) were detected [20]. A relatively low content of calcite [44] is also noticed in this, previously fired, material. Thus, based on preserved illite-mica phases and the leftover calcite in the material, it is confirmed that the roof tiles were originally fired at the factory at temperatures below 750-800 °C [45-47]. Since the same position of the peaks would be related to anorthite in a material fired at 800-830 °C, this is another proof of the low firing temperature of the roof tiles [48]. Hematite is also seen [49], which is along with the relatively high content of Fe<sub>2</sub>O<sub>3</sub> (Table 1) and FTIR. The main differences between the soils were found in the presence of clay minerals, AS contained kaolinite and illite-mica, while LS was composed of montmorillonite, illite-mica and kaolinite. Besides, the contents of hematite and calcite in LS were more pronounced compared

# 4.1.2. Firing behavior of the roof tiles waste

DTA/TGA analysis of the waste powder from 30 to  $1000\,^{\circ}$ C is shown in Fig. 4. The overall mass loss that occurs as a result of heating was around 3% for RTW. The differences between TGA and LOI results stem from the initial moisture removal by drying, absorbing the moisture from the air, and also the atmosphere in which the process took place

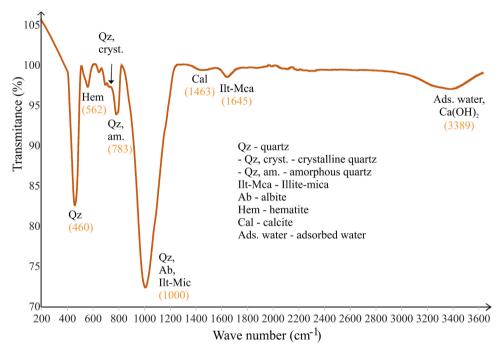


Fig. 2. FTIR result of demolished roof tile waste.

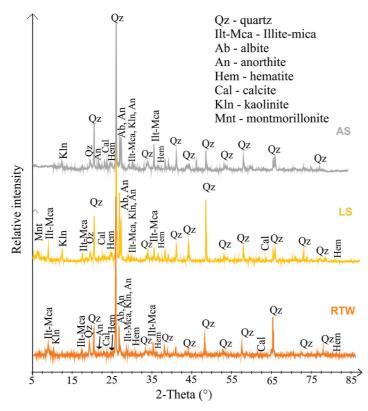


Fig. 3. XRD results of the alluvial soil (AS), laterite soil (LS), and roof tile waste (RTW) powder.

(oxygen and nitrogen). When the temperature was raised from 30 to 200  $^{\circ}$ C, weight loss was seen in the waste powder. The removal of moisture, interlayer OH-groups, and trapped carbon dioxide results in a weight loss of roughly 1.5% in the waste, which was accompanied by an endothermic reaction taking place at 165  $^{\circ}$ C [50,51]. A small amount of purely crystallized goethite is possible at about 338  $^{\circ}$ C, characterized by a small weight change and a relatively large endothermic energy effect

[52]. By further heating, there was a decrease in mass between the temperatures of 300 – 421 °C. Organic substances are released in this period [41] that accounted for less than 0.3% of RTW. At about 486 °C, an endothermic peak related to the dehydroxylation of a low leftover quantity of clay minerals was detected. Above this temperature, the exothermic reactions take place continuously up to 1000 °C. The energy released by the processes was significant enough to almost overlap the

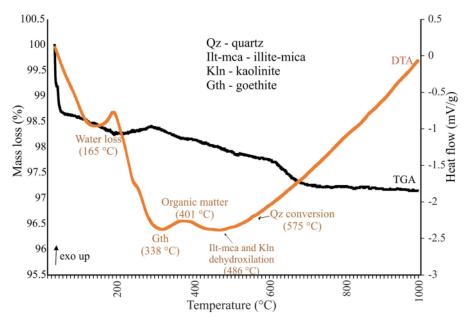


Fig. 4. DTA/TGA graph of roof tile waste.

conversion of quartz at 575  $^{\circ}$ C and to completely cover up the decomposition of some calcite left in the RTW. This effect might indicate that the roof tiles produced in the industry were fired at a low temperature, due to the ongoing reaction of Al-Si spinel and primary mullite crystallization along with a formation of the amorphous silica [53].

# 4.1.3. Micro-structural analysis

The scanning electron microscopy (SEM) image and energy-dispersive X-ray spectroscopy (EDX) analysis of demolition RTW is given in Fig. 5. The main constitutes from the image presented were O, Si and Al, and also other elements like Fe, Na, C, Ca, K, and Mg were seen (Spectrum 6–10, Fig. 5). The SEM image shows that the surface of the

material was crystalline in nature and that it was composed of Si and Al mainly as shown in the instrumental analyses (XRF, XRD, FTIR, DTA/TGA).

# 4.1.4. Toxic metal analysis

Toxicity Characteristics Leaching Procedure results are presented in Table 2. The results indicate that the limitations prescribed by the Hazardous Waste Management Rules [54] for the values of the leachates that are created from the waste of roof tiles that have been demolished were significantly lower than the values that were created by the waste. Because of this, the utilization of RTW in the production of burnt bricks results in an eco-friendly product containing non-hazardous waste.

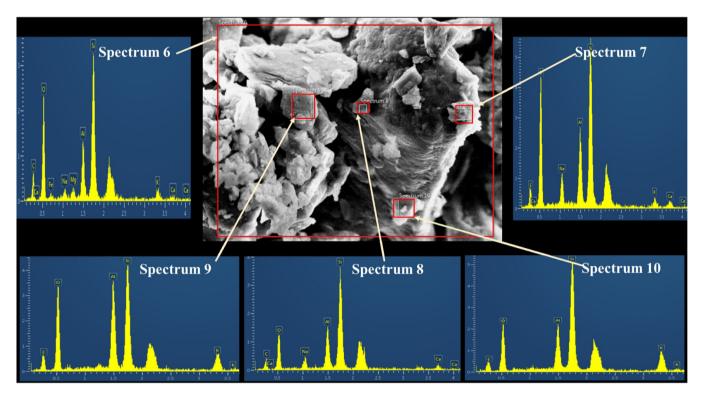


Fig. 5. Micro-structure analysis of demolished roof tile waste.

**Table 2**Toxic elements leachate in roof tile waste (RTW).

Elements	Cd	Cr	Cu	Fe	Ni	Mn	Pb	Zn
RTW (mg/kg)	2.47	0.89	0.32	0.08	0.19	2.08	0.77	1.64
TCLP hazardous waste limit (mg/kg)[57]	1.0	5.0	/	/	/	/	5.0	/

However, it must be noted that the concentrations found in leachate were significantly higher than those in natural waters that are considered contaminated [55]. The levels of heavy metals were far below those in soil sediment leachates [56]. When compared to the TCLP hazardous waste limits [57], specific caution should be paid to cadmium as the detected concentration exceeded the US EPA rules. However, using the roof tiles as a waste in some percentage of the raw clay lowers the risk of leaching.

# 4.2. Fired brick experimental results

Fig. 6. Illustrates a relatively high linear shrinkage during firing (700, 850, and 900 °C) in all the bricks. Firing shrinkage varied between 4.53% and 6.47% and was increased by raising the firing temperature and the percentual share of the RTW added. These results are influenced by the nature of alluvial and laterite soils, and especially their clay minerals content. Namely, the greater changes in the linear firing shrinkage were noticed in the case of AS in most of the cases (up to 11.9% at 700 °C, up to 17.4% at 850 °C, and up to 26.8% at 900 °C, compared to the bricks without waste addition). The reasoning behind this higher sensitivity of the AS to shrinkage is the higher content of clay minerals and organic matter which promote the densification. Besides, the AS contained more fine fractions (clay- and silt-sized) than LS [26, 28]. Furthermore, the content of fluxing oxides was increased with the addition of RTW, while filling the inter-granular spaces and inner pores, and thus an increased shrinkage with firing temperature is observed [58]. There was a correlation between the addition of the RTW and an increase in the shrinkage of the bricks across all three temperatures and both soil types (Fig. 6).

The loss on ignition of bricks that are formed by inserting waste from demolished roof tiles into both types of soils was relatively low, especially while firing at 700  $^{\circ}$ C (Fig. 7). The values of loss on ignition varied

from 5.22 to 7.02%, and increased with the rise in firing temperature, while decreased with higher share of the waste. A decrease in the values in both types of soils is caused by the addition of waste, since decreasing organic matter and clay minerals content [58]. The percentual changes in loss on ignition of the bricks of all the combinations of clays, waste and firing temperature showed that the changes were higher in the case of AS in most of the cases (up to -23.7% at 700 °C; up to -18.8% at 850 °C; and up to -17.8% at 900 °C, compared to the bricks without waste addition). The AS was the one containing more organic matter than LS, and thus the introduction of inorganic waste such as RTW introduced greater changes in loss on ignition. Besides, the higher concentration of Fe(OH)3 in the LS induced higher losses [26,28]. At a temperature of 700 °C, the loss on ignition steadily decreased in alluvial and laterite soil until 15 wt% waste was introduced into the soils; which represented a reduction of approximately 20% in alluvial and 24% in laterite soil. There was a more intensive decrease from 20 - 45 wt% addition of RTW, which also constituted a reduction in loss on ignition. On the other hand, there were two stages to the ongoing decrease in the amount of loss on ignition. The first phase of the decrease was from 5 to 30 wt% at temperatures of 850 and 900  $^{\circ}\text{C}$  on both soils with the addition of RTW. This phase accounted for 78% and 80% of alluvial, and 84% and 61% of laterite soil, respectively, of the overall reduction. The second continuous decrease phase in loss on ignition began at 35 wt% and continued until 45 wt%. This phase represented a reduction of 22% and 20% on AS and 16% and 39% on LS, respectively, from the overall reduction at both temperatures (850 and 900 °C).

Figs. 8 and 9 show the bulk density of the laboratory-scaled and commercially-sized bricks. The bulk densities obtained were relatively low compared to the usual bricks [59] but similar to those obtained in previous studies [60]. However, the molding process differences (hand or hydraulically) and granulometry of the initial materials highly influence the results [12]. Bulk densities when adding RTW were higher compared to the addition of mixed C&D waste in the same types of soils [20]. Bulk densities rose with firing temperature while showing differing behavior with increased quantities of waste. Up to 30 wt% of RTW, while firing at 850 and 900 °C, bulk densities decreased, while they later increased with the higher contents of the waste. The reasoning behind this might be that critical contents of total fluxes are obtained thus improving the density of the matrix while promoting open pores formation. This is also caused by the very low quantity of albite mineral

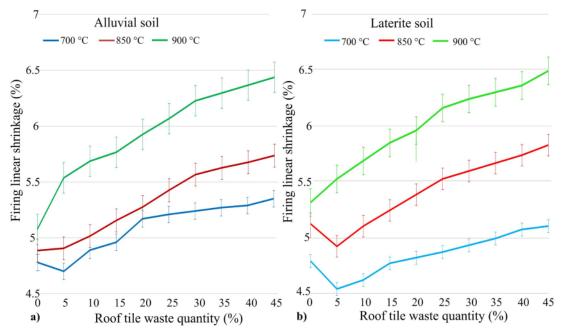


Fig. 6. Firing linear shrinkage of laboratory-scaled bricks.

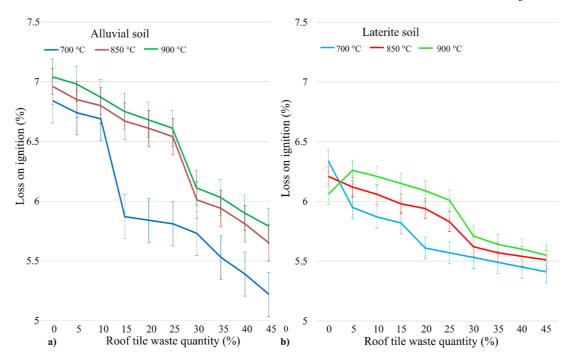


Fig. 7. Loss on ignition of laboratory-scaled bricks.

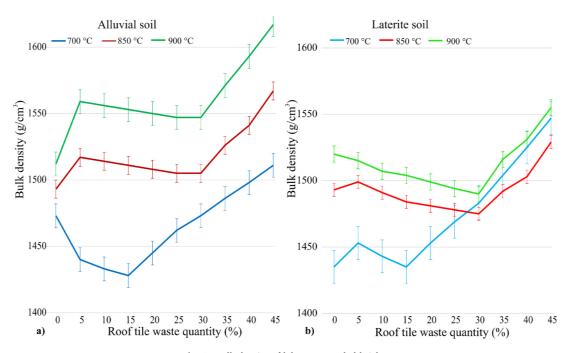
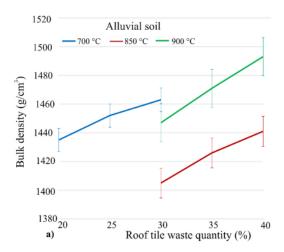


Fig. 8. Bulk density of laboratory-scaled bricks.

in the fired samples since its presence significantly lowers water absorption and open porosity [3]. The effect is also seen in the industrially-sized samples. The changes in bulk densities in the laboratory samples are mostly higher again in the AS (up to 5.0% at 850 °C; and up to 6.9% at 900 °C). After firing at 700 °C, in up to 25% of roof tile waste addition, greater changes are seen in the LS (up to +7.8%). The larger concentration of clay minerals and organic matter, and also a higher fine components concentration, in the AS were the cause of its increased changes in bulk densities [26,28]. A partly decreasing and then increasing trend of bulk density with higher incorporation of the waste was noticed also in earlier studies [58]. Bricks produced with the integration of up to 15 wt% of RTW onto AS at a temperature of 700 °C

showed a reduced bulk density on laboratory-scaled bricks. On the other hand, bulk density of the industrially-sized bricks burnt at 700 °C, showed a decline of up to 20 wt% of RTW. However, integrating RTW into AS improved the bulk density on both laboratory-scaled and commercially-sized bricks from 20 to 45 wt% and 25 to 45 wt% addition respectively. At temperatures of 850 and 900 °C, the incorporation of RTW in the soil showed a constant decline in bulk densities until the addition of 30 wt% on both laboratory-scaled and commercially-sized bricks. While a quantity ranging from 30 to 45 wt% RTW was incorporated into AS, and the bulk density of the bricks was raised at temperatures of 850 and 900 °C on laboratory-scaled and industrially-scaled brick. A 30 wt% addition of the RTW may be considered a critical



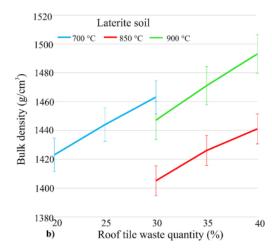


Fig. 9. Bulk density of commercially-sized bricks.

quantity that significantly changes the bulk density, especially at firing temperatures above  $850\,^{\circ}$ C. The bulk densities obtained in the industrially-sized bricks were relatively low [20], so a good thermal insulation ability is expected, which is to be further studied.

The results of the water absorption test are shown in Fig. 10. Because sintering kinetics speed up and pore capacity decreases with an increase in firing temperature, slightly lower water absorption values are obtained [58]. The maximal water absorption of the laboratory-sized bricks with the addition of RTW in AS was 26.5% at 700 °C and 24.1% at 900 °C. On the other hand, bricks manufactured with the addition of the RTW in the LS demonstrated maximum water absorption of 24.5% when heated to 700 °C, and 23.9% at 900 °C. By comparison with the bricks made of clays only, water absorption of the products containing RTW increased. The addition of RTW increased open porosity due to a little higher total quantity of carbonates in the mixtures, it improved water absorption at all three firing temperatures. Water absorption decreased with increasing the firing temperature. The changes in water absorption were higher in the AS when fired at 700 °C (up to +82.7% when 45% of RTW is added), while the LS showed higher

sensitivity with the addition of waste at 900 °C (up to +95.8% with 45% of RTW). Due to an increasing activation energy of dehydroxylation in line illite, montmorillonite, and kaolinite [61], a kaolinite-containing AS did not experience a complete dehydroxylation by firing at 700 °C, while illite, montmorillonite and kaolinite in the LS were degraded by 900 °C. By comparing the obtained results with the Indian [62] standard, up to 35 wt% of RTW can be incorporated to achieve IS 2nd class or up to 5 wt% to obtain IS 1st class bricks. Following ASTM standard [63], it is expected that an addition of 5 wt% of RTW may be used to produce 2nd class bricks after firing above 850 °C. The industrially-sized bricks showed similar results (Fig. 11). Water absorption values obtained after the addition of 35 wt% RTW satisfied the conditions for the 2nd class by Indian standard [62], while 30 wt% addition would be fulfilling the ASTM 3rd class after firing at 900 °C [63].

Bricks of laboratory scale and bricks of commercial size were used to demonstrate the compressive strength of the bricks that were produced by the integration of waste from roof tiles that had been demolished. The results of the experiments are displayed in Figs. 12 and 13. When it comes to the parameters under investigation, the percentual variations

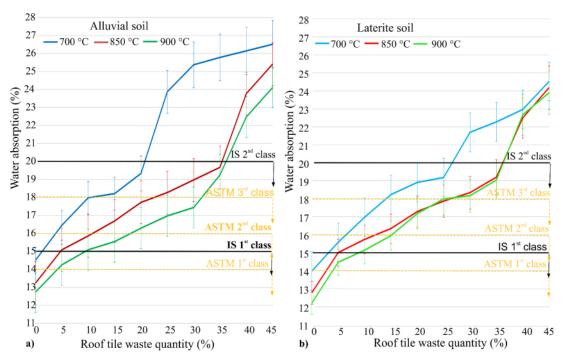


Fig. 10. Water absorption in the laboratory-scaled bricks and classes comparisons in standards in India [62] and USA [63].

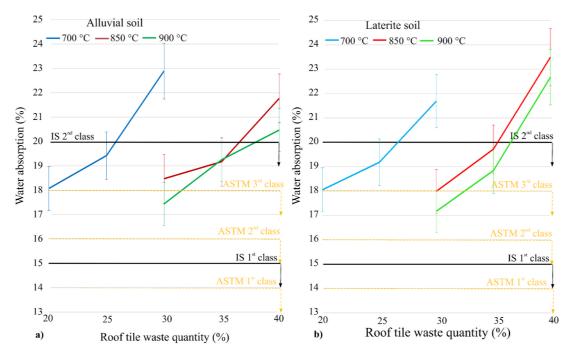


Fig. 11. Water absorption in the commercially-sized bricks and classes comparisons in standards in India [62] and USA [63].

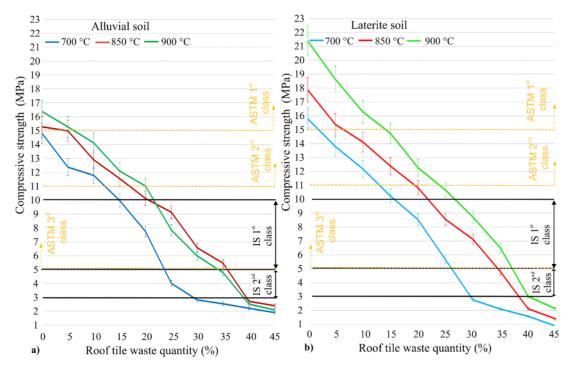
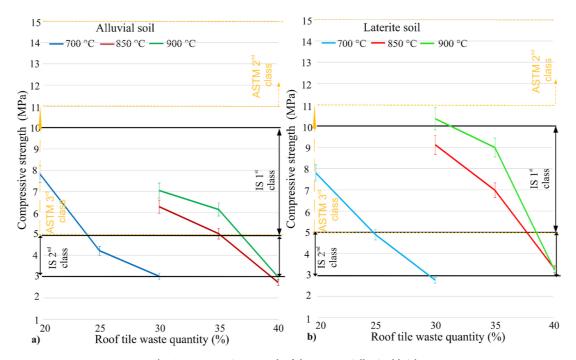


Fig. 12. Compressive strength of the laboratory-scaled bricks.

were the most pronounced when it comes to water absorption. The same effect is seen with organic waste added to these soils [27]. The compressive strength significantly drops with the addition of waste. The changes in compressive strength that are introduced with the waste were higher in the case of the LS (up to -94.5% at  $700\,^{\circ}$ C; up to -92.3% at  $850\,^{\circ}$ C; and up to -90.1% at  $900\,^{\circ}$ C). Compared to bricks made from the AS, the pure or mixed samples of LS showed a higher compressive strength. A minor amount of montmorillonite, which improved the mechanical properties of LS, is an important factor contributing to the improved particle packing (more suitable particle size distribution) [27]. The compressive strength of laboratory-scaled bricks fired at

700 °C satisfied IS 1st class at 20 wt% RTW in alluvial soil and 25 wt% in laterite soil. On the other hand, a commercial scale brick with RTW waste addition of 20 wt% met both standards (IS 1st class and ASTM 3rd class). The compressive strength of laboratory-scaled bricks fired at 850 and 900 °C satisfied the Indian (1st class) and USA standards (3rd class) for RTW addition into both soils at 30 and 35 wt% addition. On the other hand, bricks manufactured on a commercial scale with the addition of RTW met both standards (IS 1st class, 3rd class ASTM) by 35 wt% when burned at high temperatures (850 and 900 °C) for both types of soils (alluvial and laterite). [62,63].

To understand the mineralogical changes in the materials, the XRD is



 $\textbf{Fig. 13.} \ \ \textbf{Compressive strength of the commercially-sized bricks}.$ 

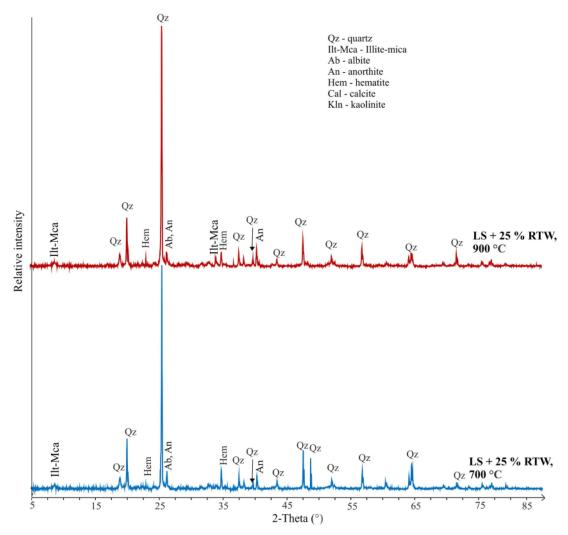


Fig. 14. Mineralogical composition of the laterite soil-based bricks containing 25 wt% of the roof tile waste (RTW) fired st 700 and 900 °C.

used to screen the samples containing 75 wt% of laterite soil and 25 wt% of the RTW (Fig. 14). This percent addition is chosen as one of the products found optimal. The fired bricks contained quartz as the dominant part [64], then the leftovers of illite-mica, albite, anorthite, and somewhat increased contents of hematite. The quantity of quartz increased after firing at 900  $^{\circ}$ C, compared to the samples fired at 700  $^{\circ}$ C. A significant portion of amorphous matter is seen as noise, especially in the sample fired at 700  $^{\circ}$ C.

## 5. Conclusions

The utilization of waste to resource is promising in the partial substitution of demolished roof tile waste (RTW) up to 45 wt% into alluvial and laterite soil for fired brick production. Based on mineralogical and chemical composition, roof tiles were determined to be initially fired in the factory below 800  $^{\circ}\text{C}$ , thus a relatively high loss on ignition at 1000  $^{\circ}\text{C}$  was determined. The Toxicity Leaching Procedure tests revealed the material exceeded the US EPA limits in terms of the concentration of cadmium, and better to be mixed as some part of the raw material for further production. The main conclusions that arose from this study are as follows:

- The incorporation of RTW caused an increase in  $SiO_2$  and a decrease in  $Al_2O_3$  contents. Besides, the RTW influenced a mild increase in  $Fe_2O_3$  concentration when alluvial soil is used and a significant decrease in the mixtures containing laterite soil,
- The introduction of RTW increased the sum of fluxing oxides and thus expected to promote better consolidation of the matrix,
- The incorporation of RTW into raw clay increased firing shrinkage and bulk density. These parameters also both increased with the temperature,
- Loss on ignition and compressive strength fell with the increase in incorporation percent of the waste, while it rose with temperature,
- Water absorption increased with waste weight percentage while decreasing with higher temperatures,
- The results related to water absorption were more limited to the quality of the product concerning the compressive strength,
- Based on laboratory examinations, respectively, up to 20 and 35 wt % of RTW can be added to produce ASTM 3rd class and Indian standard 2nd class bricks after firing at higher temperatures (850 and 900 °C),
- Based on industrial bricks testing, respectively, up to 30 wt% of RTW may be incorporated to produce 3rd class bricks after firing at 900  $^{\circ}\text{C}$  and up to 35 wt% at 850 and 900  $^{\circ}\text{C}$  to satisfy 2nd class following Indian standards,
- The fired samples containing 25 wt% of the RTW contained quartz as the dominant part, the leftovers of illite-mica, albite, anorthite, and somewhat increased contents of hematite compared to the initial mixture. The quantity of quartz increased and an amorphous matter content decreased after firing at 900  $^{\circ}\text{C}$ , compared to the samples fired at 700  $^{\circ}\text{C}$ .

This study shows encouraging research on the use of roof tile waste in fired brick production. This solution can help reduce the large amount of waste that is often dumped in unplanned areas, as well as minimize the use of natural resources for construction and building materials. Besides, considering the acceptable addition of 20-35 wt% of the waste, a carbon footprint reduction is not to be underestimated.

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# CRediT authorship contribution statement

Kalamdhad Ajay: Validation, Resources, Project administration, Funding acquisition. Laishram Boeing: Validation, Project administration, Funding acquisition. Dubale Mandefrot: Writing – original draft, Visualization, Validation, Investigation, Formal analysis, Data curation, Conceptualization. Vasic (prev. Arsenovic) Milica Vidak: Writing – review & editing, Visualization, Supervision, Data curation, Conceptualization. Goel Gaurav: Writing – original draft, Supervision, Methodology, Funding acquisition, Data curation, Conceptualization.

# **Declaration of Competing Interest**

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

# Data availability

Data will be made available on request.

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## Data Availability Statement

The data are contained within the article. Additional data are available on request.

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