



## ORIGINAL ARTICLE

# Hot mix method for the manufacture of sand-polymer bricks

## *Método de mistura à quente para fabricação de tijolos areia-polímero*

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**Abstract:** This manuscript presents a method to produce sand-polymer bricks motivated by the need to reuse plastic waste and to reduce the embodied energy and carbon footprint associated with manufacturing. The method is designed to be simple: it involves a custom-built apparatus that simultaneously mixes and heats dry sand and recycled polypropylene granules, until the latter melt. The hot mixture is then compressed in a mold until the polymer rehardens, thereby producing a bonded brick upon extrusion. Bricks with 10% and 20% polymer content (i.e., by mass,  $PC_g$ ), were prepared with either a fine sand or a coarse sand. The voids of the  $PC_g = 10\%$  bricks were approximately halfway filled with hardened polymer, whereas the voids of the  $PC_g = 20\%$  bricks were nearly entirely filled. Bricks with  $PC_g = 20\%$  were found to be potential candidates for replacement of fired clay bricks, as these exhibited infiltration levels well below specification thresholds for fired clay bricks and strengths comparable to those reported for fired clay bricks. Furthermore, the embodied energy associated with sand-polymer bricks was calculated to be around a third of that required for fired clay bricks. In addition, the manufacture of sand-polymer bricks requires minutes, whereas that of fired clay bricks requires hours. The results gathered suggest that bricks produced by the proposed method have potential for replacement of fired clay bricks in applications wherein replacement is favorable.

**Keywords:** composite, plastic, polymer, sustainability, waste.

**Resumo:** Este manuscrito apresenta um método para produzir tijolos areia-polímero motivado pela necessidade de reutilizar resíduos plásticos e reduzir a energia incorporada e a pegada de carbono associada à fabricação. O método foi pensado para ser simples: envolve um aparelho customizado que mistura e aquece simultaneamente areia seca e polipropileno granular reciclado, até que estes derretam. A mistura quente é então comprimida num molde até que o polímero endureça novamente, produzindo assim um tijolo conformado após extrusão. Tijolos com teor de polímero de 10% e 20% (isto é, em massa,  $PC_g$ ), foram preparados com areia fina ou areia grossa. Os vazios dos tijolos  $PC_g = 10\%$  foram preenchidos aproximadamente até a metade com polímero endurecido, enquanto os vazios dos tijolos  $PC_g = 20\%$  foram quase totalmente preenchidos. Descobriu-se que os tijolos com  $PC_g = 20\%$  são candidatos potenciais para a substituição dos tijolos de barro cozidos, uma vez que apresentavam níveis de infiltração bem abaixo dos limites de especificação para tijolos de barro cozidos e resistências comparáveis às relatadas para tijolos de barro cozidos. Além disso, a energia incorporada associada aos tijolos areia-polímero foi calculada em cerca de um terço daquela necessária para os tijolos de argila cozidos. Além disso, a fabricação de tijolos areia-polímero leva minutos, enquanto a de tijolos de argila cozida leva horas. Os resultados obtidos sugerem que os tijolos produzidos pelo método proposto têm potencial para substituir os tijolos de barro cozido em aplicações onde a substituição é favorável.

**Palavras-chave:** compósito, plástico, polímero, sustentabilidade, resíduo.

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**Data Availability:** The data that support the findings of this study are available from the corresponding author, [LM], upon reasonable request.

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## 1 INTRODUCTION

The rate of plastic waste production is increasing at a concerning rate. In the US, the total volume of plastic waste within solid waste grew from 0.5% to 12.5% between 1960 and 2010 [1]. Globally, estimations reveal that 75% of all the plastic ever produced has become garbage, with one third of such of plastic (100 million tons) taking part as land or marine pollution [2]. An additional ~100 million tons are forecasted by 2030 if considerable measures are not implemented [2]. There is thus a pressing urgency to find alternative strategies for reuse of waste plastics.

The production of traditional civil engineering materials (e.g., concrete, steel) requires large amounts of energy, and generates pollution [3]. For example, Portland cement manufacturing consumes 10.5 EJ of world energy [4], and the emission of a significant percentage of polluting and greenhouse gases [5]. Therefore, the search for alternative and novel -particularly recycled- materials to reduce costs and environmental impacts associated with construction has increased in the last number of years [6], [7]. Indeed, buildings and pavements have been the target of incorporating a diverse gamma of materials to improve performance and reduce construction costs [8]–[11]. Furthermore, the development of modern civil engineering construction has generated a strong demand for production of alternative materials and materials that incorporate waste products [12]–[17].

The current study is motivated by environmental problems associated with waste plastics, and by the simultaneous desire to develop sustainable materials for use in construction. In the current study, sand and recycled polypropylene, an essentially ubiquitous waste plastic, are mixed to produce solid composites; herein called sand-polymer bricks. These bricks are manufactured via heat-treatment, whereby the heated, softened polymer adheres to the mineral sand grains, producing a solid upon cooling. The authors have produced this type of polymer-bonded material earlier [18]. The current work emphasizes a distinction in the means by which the resulting brick is cast. Formerly, the mixed materials were placed in a mold and heated (in the mold) inside a convection oven or a solar oven [19]. Such method is beneficial for preparing small bricks (~2cm) but becomes increasingly inefficient as the brick size increases. This inefficiency is associated with the difficulty for heat to transfer from the brick's boundaries to the brick's core, resulting in heterogeneous softening of the added polymer. In the current work, the material is mixed and heated simultaneously in a simple, yet robust device, and then compressed in a mold while it cools. The heating-while-mixing protocol enables homogeneous softening of the polymer, and therefore, relatively large bricks of high strength can be produced rather quickly. In addition, we seek to remove as much plastic as possible from the waste stream by placing it, i.e., storing it, within these bricks, yet while not replacing any mineral grains with polymer. The use of phase relationships allows for estimation of the amount of polymer needed to produce a brick that features a dense assembly of interconnected mineral grains with low voids volume.

## 2 PHASE RELATIONSHIPS

Consider a dry mixture of plastic granules and sand grains, in a bucket. The plastic granules melt upon heating and harden upon cooling. Inspection of the hardened composite may reveal either an interconnected assembly of sand grains with polymer in its pores (i.e., voids) or a polymer matrix with suspended, non-interconnected sand grains. Furthermore, the composite may contain sites where one would find the former and sites where one would find the latter. Indeed, the microstructure of such a composite depends on the mixture fraction (i.e., polymer content) and is affected by the casting conditions. In the example presented, the mixture is simply heated inside a bucket. The microstructure of the resulting composite is thus largely unpredictable because there is no control of the mechanism by which the mixture is cast.

Previous studies associated with heat-induced polymer bonded sands emphasize strength gain with minimal polymer contents. In a composite with low gravimetric polymer content (e.g., 1%) [20] and relatively small polymer particles, i.e.,  $d_{\text{poly}} \ll d_{\text{sand}}$ , the polymer volume is much smaller than the volume of voids ( $V_p \ll V_v$ ), and therefore, the polymer particles rest initially in the voids comprised by the sand grain assembly. Upon heating, the polymer particles melt, and upon cooling, the porous composite features hardened polymer menisci at contacts. Given the low polymer content used in such studies, composites can be prepared by heating the mixed material under self-weight, i.e., with no added stresses. The current study considers the ideal filling of all voids with polymer. To create such a composite, the mixture is not heated inside a mold or bucket (as described earlier), but instead, the mixture is compressed while the plastic is molten, so that it hardens while the assembly of mineral grains has a high density.

The heat-induced polymer bonding of aggregates translates to a unique set of phase relationships because the polymer is a solid before and after, yet a fluid during the heat treatment. While hot, the relevant phases are thus air, liquid polymer, and solid sand grains. Both air and the polymer may occupy void volume, assuming that the mineral grains comprise an interconnected assembly. Thus, the fraction of polymer that occupies the void space of the mineral grain assembly can be defined as

$$F_p = \frac{V_p}{V_p + V_a} \tag{1}$$

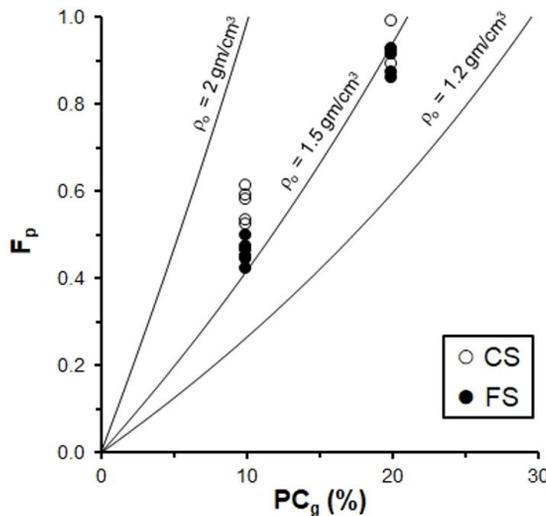
where  $V_a$  is the volume of air.  $F_p$  can be written as a function of the gravimetric polymer content, i.e.,

$$PC_g = \frac{m_p}{m_p + m_{mi}} \tag{2}$$

using density definitions, i.e.,  $\rho_p = m_p/V_p$ ,  $\rho_{mi} = m_{mi}/V_{mi}$ , and  $\rho_o = (m_{mi})/(V_a + V_p + V_{mi})$ , where  $m$  stands for mass and the subscript ‘mi’ stands for mineral, i.e.,  $\rho_{mi}$  is the density of the mineral that the soil grains are made of. The density  $\rho_o$  contains only the mass of the soil grains (i.e., excludes the polymer mass). Thus,  $\rho_o$  represents a desired density for the soil grain assembly, for example, the Proctor dry density of the candidate soil. Writing Equation 1 in terms of Equation 2 yields

$$F_p = \frac{PC_g \rho_o \rho_{mi}}{\rho_p (PC_g \rho_o - PC_g \rho_{mi} - \rho_o + \rho_{mi})} \tag{3}$$

The three curves in Figure 1 (disregard the data points for now) shows the variation in  $F_p$  as a function of  $PC_g$  for three values of  $\rho_o$ , which represent a common range of maximum Proctor dry densities for sandy soils [21], computed with Equation 3 and  $\rho_p = 0.92 \text{ gm/cm}^3$  (density of the polymer used in the current study).



**Figure 1.** Fraction of polymer that occupies the void space ( $F_p$ ) versus gravimetric polymer content ( $PC_g$ ).

Clearly, the polymer content required to fill the voids decreases as  $\rho_o$  increases, as there is decreasing voids volume for the polymer to fill.

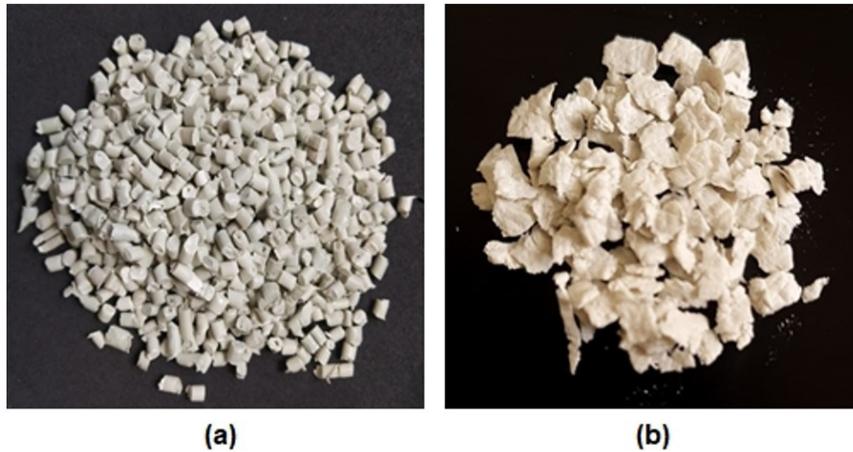
The porosity of the hardened composite is

$$n = \frac{V_a}{V_a + V_p + V_{mi}} \tag{4}$$

The ideal composite would be one where  $n = 0$  and  $F_p = 1$ , provided that the mechanical properties of the composite are appropriate for the application in question. If the composite is a construction brick, then the material benefits from having a dense mineral grain assembly (for strength), and the maximum amount of polymer stored in its voids (for removal of plastic from the waste stream, and for low water absorption).

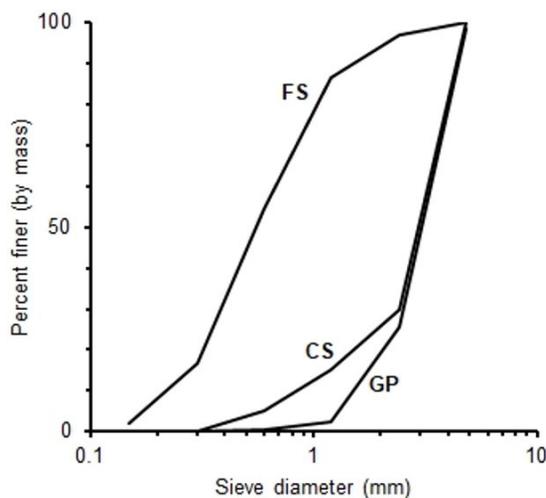
### 3 MATERIALS AND EXPERIMENTAL PROGRAM

The recycled polypropylene was made available by the company PLAST MAN in the form of uniform ( $d_p \approx 5$  mm) granular pellets with specific gravity  $G_s = 0.92$  (Figure 2a), which were consequently ground into smaller grains (Figure 2b) using a knife mill (TECNAL). This was done simply by feeding the pellets continuously into the funnel of a grinding mill. The resulting ground polypropylene (GP) with coefficient of uniformity  $C_u = 2.6$ , was used for the preparation of mixtures. The tensile modulus and tensile strength of polypropylene are in the range of 1.9 GPa and 38 MPa, respectively [22].



**Figure 2.** Pictures of granular polypropylene before (a) and after being grounded (b). Image (a) is  $\sim 5$  mm across. Image (b) is  $\sim 3$  mm across.

The grain size distribution (GSD) curve for GP is shown in Figure 3. Two different sands were selected to explore the role of relative grain sizes on the strength and water absorption properties of the prepared bricks: a fine sand “FS” with  $G_s = 2.65$  and grain sizes considerably smaller than those of the GP, and a coarse sand “CS” with  $G_s = 2.65$  and a mean grain size similar to that of the GP. GSD curves are shown in Figure 3. The FS ( $C_u = 3.45$ ) was obtained from the Taperoá river in Paraíba, Brazil. The CS ( $C_u = 3.89$ ) was obtained from a rock quarry in Campina Grande in Paraíba, Brazil. It is worth noting that CS is ‘stone dust’, i.e., waste sand grains produced during rock crushing for production of aggregates [23]. Currently, stone dust has little to no commercial market value and is therefore often stocked in quarry yards that form large waste piles. The potential use of stone dust for the development of new construction materials, as described herein, can therefore offer a possibility for the reduction of the costs associated with its storage.



**Figure 3.** Grain size distribution curves of materials used

Each mixture was manufactured as follows. A predetermined mass of sand is first mixed dry manually with a predetermined mass of GP, such that a desired polymer content, either  $PC_g = 10\%$  or  $PC_g = 20\%$ , is attained. These polymer contents were selected with guidance from Equation 3, assuming that the sands utilized would approach an ultimate mass density  $\rho_o = 1.5 \text{ gm/cm}^3$ . For  $PC_g = 10\%$ , Equation 3 yields a predicted  $F_p$  of about 0.5 (i.e., the polymer occupies half of the voids space), and for  $PC_g = 20\%$ , Equation 3 yields a predicted  $F_p$  of about 1 (i.e., the polymer fills most, if not all the voids volume).

#### 4 APPARATUS

Each composite was manufactured utilizing a custom-made apparatus inspired by a manual injection molding machine. Sketches are shown in Figure 4.

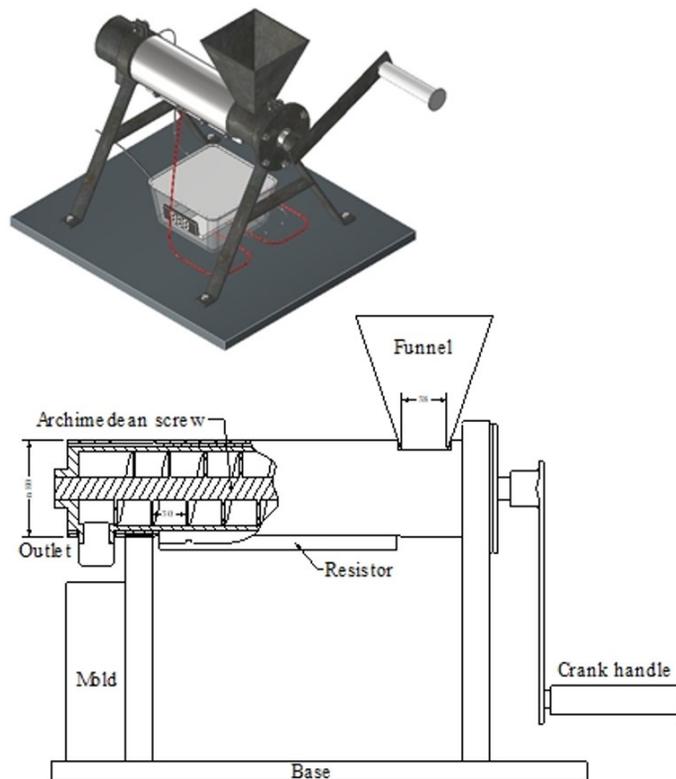


Figure 4. Scaled schematics of the custom-made apparatus.

The apparatus is composed of a horizontal chamber equipped with a temperature controller, temperature sensor, a controller activation key, a collar-type electrical resistor, and a hand crank. The cold polymer-soil mixture enters the apparatus through a top funnel. The material falls by gravity into the horizontal cylindrical chamber, below, which houses an Archimedean screw. The chamber is wrapped by the electrical resistor and contains a temperature sensor; both are connected to an Arduino system that sustains the temperature inside the chamber at  $170 \text{ }^\circ\text{C}$ . The Archimedean screw mixes, heats, and transports the material in response to the operation of the hand crank. On average, the hand crank is rotated such that any portion of the mixture traverses the 35 cm long chamber in about 15 minutes. Such time, found by trial and error, is the minimum duration required to render sufficient heat to cause melting of the plastic as the material is transported through the apparatus. The hot mix exits the chamber and falls by gravity into a cylindrical steel mold with diameter = 5 cm. Pre-lining of the mold with wax paper or aluminum foil, as in previous studies [19], [20], was not necessary because polypropylene does not adhere readily to steel. The mixture is allowed to cool in the mold under a sustained stress of approximately 600 kPa using a hydraulic press. This compression stage promotes improvements in the composite through better adhesion onto the grains as well as air expulsion. Only 5 minutes are needed for the mixture to cool into a solid body. Once cooled and hardened, a ~10 cm long cylindrical sand-polymer brick is extruded from the mold with the press. A brick with  $PC_g = 20\%$  is shown in Figure 5.



Figure 5. Polymer bonded CS cylinder with  $PC_g = 20\%$ .

## 5 BRICK TESTING

Infiltration tests were carried out following the guidelines of ABNT NBR 9778 [24], as follows. First, the mass of each brick is measured. Second, each brick is immersed in filtered tap water at room temperature. Third, each brick is removed from submersion after 24 hours, whereby its mass is determined, followed by resubmersion. The mass of each brick is also measured 48 and 72 hours into the submersion duration. Fourth, each brick is then removed from the water bath and left to dry completely. Finally, each brick is subjected to either a tensile strength via diametrical compression test following ABNT NBR 7222 [25], or a compression test following ABNT NBR 7215 [26]. In the former, the tensile strength is the tensile resistance at the onset of diametrical fracture.

## 6 RESULTS

### 6.1 Polymeric fraction

The  $F_p$  values for the manufactured bricks are shown in Figure 1 where each data point corresponds to a brick. Note that a relatively high level of polymer filling was attained for  $PC_g = 20\%$  bricks, i.e.,  $F_p \approx 90\%$ , whereas the  $F_p$  values for  $PC_g = 10\%$  bricks are close to 50%; both reasonably predicted with Equation 3.

### 6.2 Water absorption

The evolution of the water absorbability, i.e.,  $w = m_w / (m_{mi} + m_p)$  = mass of water/mass of solids, the degree of saturation, i.e.,  $S = V_w / V_v$  = volume of water/volume of voids, and the permeable porosity, i.e.,  $n_p = V_w / V =$  volume of water/total volume, are shown in Figure 6.

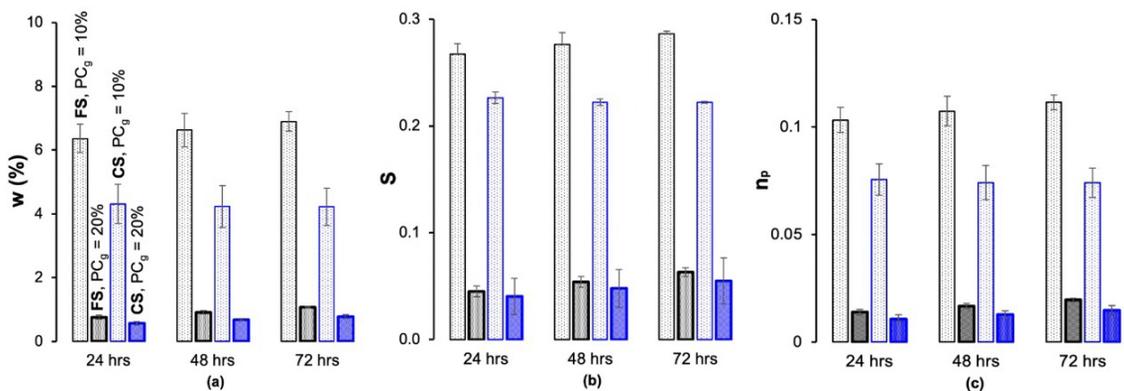


Figure 6. Evolution of water absorbability,  $w$  (a), degree of saturation,  $S$  (b), and permeable porosity,  $n_p$  (c), for bricks submerged in water.

The following observations can be made. First, the water absorbability for all bricks (Figure 6a) is considerably lower than the maximum permitted by American and Brazilian standards for fired clay bricks, i.e., 20% [27]; this is particularly so for  $PC_g = 20\%$  bricks for which  $w < 1\%$ . Indeed, typical values for fired clay bricks range between  $w = 14$  and  $16\%$  [28], [29]. Second, bricks do not attain full saturation (i.e.,  $S = 1$ ; Figure 6b). Indeed, the maximum level of saturation measured is only  $< 30\%$ , as most of the pores are closed ( $n_p \leq 0.1$ ; Figure 6c). Third, bricks composed of FS absorb more water than analogous bricks composed of CS. A possible explanation is that in FS bricks, a larger amount of mineral surface area is left untouched by the polymer, and therefore, water infiltrates the FS brick more easily along unbonded regions. Fourth,  $w$ ,  $S$ , and  $n_p$  decrease substantially as  $PC_g$  increases: water infiltration is reduced significantly as the content of the impervious polymer increases. Fifth, the rise in  $w$ ,  $S$ , and  $n_p$  associated with time is negligible, as most of the ultimate level of saturation is reached only after 24 hrs. This means that the pervious paths contained in each brick, albeit small, are readily infiltrated by water.

### 6.3 Tensile and compressive strengths

The tensile strengths of tested bricks are shown in Figure 7a as a function of the  $F_p$  (Equation 3), and in Figure 7b as a function of the porosity,  $n$  (Equation 4). Figure 8 shows the compression strengths of tested bricks in the same format as Figure 7.

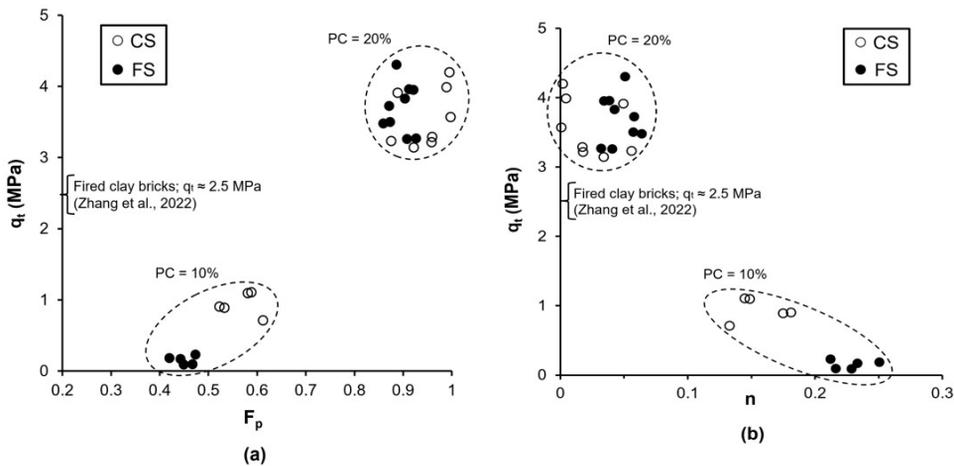


Figure 7. Tensile strength of tested bricks as a function of polymer fraction (a) and porosity (b).

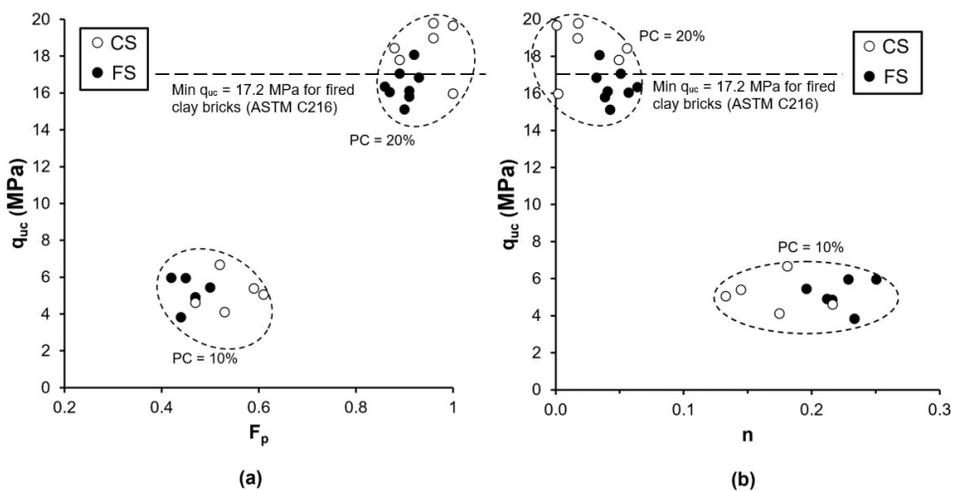


Figure 8. Compressive strength of tested bricks as a function of polymer fraction (a) and porosity (b).

Figures 7 and 8 show that both  $q_t$  and  $q_{uc}$  increase with increasing  $PC_g$ . A salient result is that the tensile strengths of the  $PC_g = 20\%$  bricks, i.e.,  $q_t = 3.2$  to  $4.2$  MPa, are considerably higher than those reported for common fired clay bricks quasistatically-loaded in split-tension (i.e.,  $q_t \approx 2.5$  MPa) [30]. Similarly, compression strengths, i.e.,  $q_{uc}$  values are within the range of the minimum permissible per ASTM C216 [31] for fired clay bricks ( $q_{uc} = 17.2$  MPa); and in particular, above such minimum for CS bricks.

Recall that when compared to CS, FS has a relatively large surface area relative to the amount of available polymer. Therefore,  $F_p$  is lower for FS bricks than for CS bricks, for the same polymer content (Figures 1, 7, and 8). The role of surface area on strength is most evident via the difference in tensile strength between FS and CS bricks with  $PC_g = 10\%$ , as shown in Figure 7a, for example. The tensile action imposes pulling of the polymer bridges that bond mineral grains as well as forced opening of voids. For FS bricks with  $PC_g = 10\%$ , the small amount of polymer relative to the large surface area results in resistance offered by a relatively small number of polymer bridges, and therefore small  $q_t$  values.

The strength ratio  $q_{uc}/q_t$  is commonly reported for engineering materials. The average strength ratio for bricks with  $PC_g = 20\%$  (i.e., high) was found to be is  $\sim 5$  regardless of sand type. By contrast, the strength ratio was found to be dependent on sand type for bricks with  $PC_g = 10\%$  (i.e., low), i.e.,  $\sim 35$  for FS and remaining at  $\sim 5$  for CS. The high value for FS (vs. that for CS) suggests that for low polymer contents, the mechanisms associated with tensile resistance (i.e., pulling action and opening of voids) are highly sensitive to soil's mineral surface area relative to the available polymer. By contrast, the mechanisms associated with compression (pressing and closing of voids) appear to be unaffected by surface area when the polymer content is at least 10%.

## 7 EMBODIED ENERGY AND ENVIRONMENTAL CONSIDERATIONS

Material replacement is practically viable when environmental and/or energy costs are smaller than those associated with the material being replaced [32], [33]. In particular, the embodied energies associated with the manufacture of both fired clay bricks and the sand-polymer bricks presented herein are largely governed by heating. The energy required to heat a body from  $T_1$  to  $T_2$  is  $E = \Delta T_{pc}$ , where  $\rho$  is the mass density,  $c$  is the specific heat capacity, and  $\Delta T = T_2 - T_1$ . The following approximations can be made. For fired bricks,  $E \approx 2000 \text{ kg/m}^3 \cdot 800 \text{ J/(kg}^\circ\text{C)} \cdot (900^\circ\text{C} - 20^\circ\text{C}) = 1400 \text{ MJ/m}^3$ . For sand-polymer bricks,  $E \approx 1900 \text{ kg/m}^3 \cdot 1400 \text{ J/(kg}^\circ\text{C)} \cdot (170^\circ\text{C} - 20^\circ\text{C}) = 400 \text{ MJ/m}^3$ . Therefore, sand-polymer bricks could be produced using only  $400/1400 \approx 1/3$  of the energy used to produce fired bricks.

In addition to savings in embodied energy, sand polymer bricks do not require use of water (as do fired clay bricks). The replacement of water and cement by waste polymeric materials also translates to a reduction of greenhouse gases and pollutants [3]–[5].

Polymers have the propensity of releasing volatile organic compounds (VOC) as they degrade. Polypropylene, the polymer utilized herein, does not release VOCs upon degradation below  $200^\circ\text{C}$ , whereas it does generate olefinic compounds above  $200^\circ\text{C}$ , with their quantity increasing with temperature [34]. At much higher temperatures, i.e., above  $330^\circ\text{C}$ , the main gaseous products obtained from the thermal degradation of polypropylene/biomass composites are  $\text{H}_2\text{O}$ ,  $\text{CO}_2$ ,  $\text{CO}$ , formaldehyde, methanol, acetic acid, formic acid, and methane [35]. Such knowledge is encouraging in that the degradation of polypropylene does not generate collectable amounts of VOCs at  $170^\circ\text{C}$ , i.e., the temperature imposed to manufacture sand-polymer bricks with the method described herein.

## 8 CONCLUSIONS

A readily implementable, water-less method for the manufacture of bricks composed of sand and plastic, motivated by the need to remove plastics from the waste stream, is presented. The raw materials are sand and granulated recycled polypropylene. The method involves the use of a custom-made apparatus that transports, heats, and mixes the material via an Archimedean screw. The apparatus releases the mixed material, which contains molten plastic, into a mold wherein the mixture is compressed until the plastic rehardens upon cooling. The extruded composite is a bonded brick.

Results of experiments indicate that bricks prepared with a polymer content of 20%, which produced nearly complete void filling when mixed with the sands utilized, feature strengths and water infiltration levels that are more favorable or at least comparable than those attributed to common fired clay bricks. Furthermore, the embodied energy calculated for the former is approximately one third of the latter; and the production time of the former (minutes) is substantially lower than that of the latter (hours). Such findings offer encouragement towards the possibility of utilizing the described method to produce candidates for fired brick replacement in favorable applications.

Phase relationships, particularly Equation 3, can be used to predict the gravimetric polymer content required to maximize the filling of the mineral assembly's pore space with plastic, provided that the polymer's density  $\rho_p$  and soil's target density  $\rho_o$  (for example, Proctor) are known.

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