

Manufacturing and Properties of Gypsum-Based Products with Recovered Wood and Rubber Materials

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The experimental production of gypsum-based products (cylindrical samples, solid bricks) using different fractions of wood chips and rubber particles was studied. Recovered rubber and wood materials were mixed with gypsum and water in various proportions to fabricate gypsum-wood and gypsum-rubber cylindrical samples and standard solid bricks with six holes using appropriate molds. It was shown that to manufacture gypsum-wood and gypsum-rubber products with good mechanical strength, coarse fractions of wood and rubber should be used, but the proportion of wood or rubber should not exceed 25%. No thermal conductivity differences were found between the wood- and rubber-type of gypsum products, and particle size and material proportion had no effect. Samples with fine wood and rubber particles present at a lower proportion (25%) exhibited similar sound absorption behavior. The solid bricks had slightly higher strength when loaded at the large surface of their lateral upper side than when loaded at the small surface. The bricks provided better thermal insulation than both the extruded and pressed house bricks but lower than that of insulating bricks. The emission of volatile organic compounds out of the bricks was at an acceptable level according to regulations for construction products.

Keywords: Wood chips; Rubber particles; Solid bricks; Compression; Thermal conductivity; Sound absorption; VOCs

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INTRODUCTION

With the world's increasing population and wealth, recycling has become vital for both environmental health and society. There is growing interest in reducing expenditure by making products from recycled materials. Doing so not only preserves natural resources and requires less energy but also is preferential to many consumers who desire products with a lower environmental impact (Essoussi and Linton 2008). The policies of most countries are evolving to encourage continued growth in recycling and the use of recycled materials, and often specific recycling rate targets apply for different waste fractions (European Union 2008).

Waste tires are a major concern because the number of them is increasing due to the improvement in the worldwide standard of living and the short lifetime of tires. It is estimated that each year in the 27 EU Member States and Norway, over 300 million tires are permanently removed from cars and discarded as waste (ETRA 2013). Today, these post-consumer tires are valorized in different ways and are increasingly being considered

valuable resources. Material recycling and energy recovery are the two primary means of valorization and result in an array of sustainable materials useful in many applications, particularly within the construction sector. During the past two decades, accumulated used tires in the EU Member States totaled approximately 50 million tons; 10 million tons of these accumulations were materially recycled. At the same time, the disposal of post-consumer tires has caused environmental and economic problems to many countries, and environmental concerns regarding their incineration have risen during the last few years (Stefani *et al.* 2005). It is expected that the EU, given its goal of increasing recycling and eliminating landfilling, will place additional demand on EU tire recyclers in the future and on the research community to develop new methods to recycle waste tires.

Waste wood is another valuable secondary raw material that fulfills much of the wood-based panel industry's needs and serves as a major source of renewable energy. A very large fraction of waste wood still ends up in landfills or is composted (Merl *et al.* 2007). Much waste wood comes from households (furniture, doors, windows, floors, and the like) and production residues from wood industries. These waste wood fractions are contaminated by chemicals including lacquers, paints, coatings, and adhesives (mainly formaldehyde-based) or preservatives, making their recovery problematic. Several methods have been proposed, especially for the recycling of waste wood-based panels (particleboards, fiberboards), including mechanical handling, steaming, heat treatment, pulping, and hydrothermal treatment (Moeller 1993; Roffael 1996; Hesch 2002; Riddiough 2002; Sandison 2002; Michanickl and Boehme 2003; Lykidis and Grigoriou 2008). In the future, waste wood is expected to play an increasingly important role in the sustainability of wood industries and in the protection of the environment. Therefore, new technologies are needed to fully explore this valuable resource.

Waste tire rubber is a promising raw material for use in composites, as it is lightweight and has good elasticity, energy absorption, insulating (both sound and heat), anti-caustic, and anti-rot properties (Fu 2003). The feasibility of manufacturing wood-rubber composite panels using different binder systems (*e.g.*, commercial resins), has been studied previously. The mechanical properties of particleboards made using melamine-urea formaldehyde and polyisocyanate were found to comply with the general-purpose particleboard minimum property requirements of EN 312 Type P1 (2005) at 10% waste tire rubber loading, and the tire rubber improved the water resistance of the particleboards due to its hydrophobicity (Ayrilmis *et al.* 2009). Jun *et al.* (2008) suggested some optimal conditions (pressing temperature, pressing time, and board density) for the manufacture of wood-rubber functional composite panels with a combination polymeric methylene diphenyl diisocyanate and urea-formaldehyde binder system. Agricultural lignocellulosic materials (rice straws) combined with rubber have also been studied for use in insulation boards manufactured using a commercial polyurethane adhesive as the composite binder (Yang *et al.* 2004). It was found that the water proof, water absorption, and thickness swelling properties of the rubber composite boards were better than those of wood particleboard, and that the flexibility and flexural properties were superior to those of other wood-based panel products. They also demonstrated good acoustic insulation, electrical insulation, anti-caustic, and anti-rot properties.

Due to its availability, relative low cost, and easy handling, gypsum is widely used as a construction material, especially for nonstructural components such as gypsum wall board and ceilings (Goodall and Gupta 2011). Reinforcing materials such as polymeric fibers are usually added to the gypsum board to improve certain mechanical properties (Yu and Brouwers 2012; Gencel *et al.* 2014). Gypsum composites reinforced with waste fibers

such as recycled cellulose pulp have also shown to be a technically better substitute for the brittle gypsum board, with higher impact strength and modulus of rupture (Carvalho *et al.* 2008). Bricks belong to the wide family materials for the construction of outer and inner walls in buildings, and various attempts were made to incorporate various waste materials in the brick industry such as wood sawdust and cotton (Turgut 2007; Algin and Turgut 2007). Gypsum has been investigated little as an inorganic binding material for the manufacturing of bricks from various industrial and agricultural wastes (Raut *et al.* 2011), but could open new possibilities in converting waste into useful building and construction products. Investigating the effects of various waste materials to be used on the physical and mechanical properties of gypsum-based bricks as well as their thermal insulation would be essential, as it is the case for similar products (Alaa *et al.* 2013). An advantage of using natural fibers in composite products and bricks is their high energy-absorbing capacity due to their low modulus of elasticity, and composites containing waste fibers have presented good mechanical performance compared to composites reinforced with virgin fibers (Savastano *et al.* 2005).

Promoting sustainable raw material management and increasing the recycling of waste materials are two strategies to opening large domains for new technologies and innovations. The present study was aimed to increase resource use efficiency through the utilization of wood and rubber wastes for the manufacture of a new construction product. The manufacture and testing of gypsum-based samples and solid bricks with wood chips from particleboard production residues and rubber from waste tires were investigated. The proposed products could be alternatives over other, traditional, less eco-efficient materials currently used in the residential construction sector.

EXPERIMENTAL

Fine (0.2 to 0.8 mm) and coarse (2 to 4 mm) rubber particles as well as textiles from tire recycling were provided by the Greek companies Keridis Christoforos SA, Arabissos, and Retire SA, Drama. Wood chips from particleboard production residues were also purchased in two fractions, fine (0.4 to 1.25 mm) and coarse (3.15 to 4 mm), from Glunz AG, Germany. Additionally, a medium wood fraction (1-2 mm) was prepared from the coarse fraction after sieving (Fig. 1). The recovered materials (rubber, wood) were fully mixed manually within 10 min with gypsum and water and the mixture was poured into cylindrical molds and left to air-dry. For comparison, different gypsum/wood/rubber ratios for each fraction (fine, coarse) were used, as well as different gypsum/water ratios (Table 1). Two categories of gypsum-based cylindrical products were manufactured, gypsum-rubber and gypsum-wood, while pure gypsum samples served as controls.

Gypsum-based solid bricks were manufactured using the following raw materials and proportions per weight: (a) 15% medium-sized fraction (1 to 2 mm) wood chips; (b) 10% of a coarse fraction (2 to 4 mm) rubber; (c) 5% rubber-textile as a reinforcement material; and (d) 70% gypsum (Fig. 1). The gypsum/water ratio was 1:1.5. The wood chips and rubber particles were purchased from the same companies previously mentioned for the manufacture of the gypsum-based cylindrical samples. After manual mixing of materials in plastic bowls within 10 min, the mixture was poured in special rectangular molds with selected standard final brick dimensions of $85 \times 55 \times 185 \text{ mm}^3$ (W \times T \times L) and six symmetrical round holes. After the production process, the bricks were air-dried at laboratory conditions.



Fig. 1. Fine (a) and coarse (b) rubber particles, rubber-textile (c), and fine (d), medium (e) and coarse (f) wood chips. The rubber fractions (a) and (b) and the wood fractions (d) and (f) were used for cylindrical samples. The rubber fractions (a) and (b), the rubber textile (c) and the medium wood fraction (e) were used for production of bricks

Table 1. Manufacturing Details of Gypsum-based Cylindrical Products (wt%)

Product Code	Gypsum (G)	Rubber Particles (R)		Wood Particles (W)		Gypsum/ Water Ratio
		Coarse (c)	Fine (f)	Coarse (c)	Fine (f)	
G	100					1:1
GR _{c25}	75	25				1:1
GR _{c50}	50	50				1:1
GR _{f25}	75		25			1:2
GR _{f50}	50		50			1:3
GW _{c25}	75			25		1:1
GW _{c50}	50			50		1:3
GW _{f25}	75				25	2:3
GW _{f50}	50				50	2:3

All gypsum-based products (cylindrical and solid bricks) were tested for their compressive strength, thermal conductivity, and sound absorption (Table 2). Testing of the thermal conductivity and sound absorption of the solid bricks at the lateral direction required appropriate machining to produce cylindrical samples. Compression testing of the cylindrical products and solid bricks was performed with a Shimadzu UH-300 kNI (Japan) testing machine using ASTM standards C39/C39M-12a (2014) and D1037-12 (2012), respectively. In the case of the cylindrical products, the axial compression was determined, while the solid bricks were tested for their compression strength in the axial and two lateral directions.

Table 2. Samples and Standards Used for Testing of Gypsum-based Cylindrical Products and Solid Bricks

Property	Number of Samples	Dimensions (mm)		Standard
		Diameter	Height	
Compressive Strength*	20	50	100	ASTM C39/C39M-12a
Thermal Conductivity	10	50	20	ASTM E1530-11
Sound Absorption	10	100	50	ISO 10534-1

* In the case of solid bricks, orthogonal samples 100 mm in height were used for the axial compression (surface 55 × 85 mm²), and samples 85 × 85 × 55 mm³ (W × T × L) and 85 × 55 × 85 mm³ were used for the lateral compressive strength for loading perpendicular to their large and small lateral surfaces, respectively. The respective standard used was ASTM D1037-12 and the sample size was 20 for each direction.

The thermal conductivity coefficient (k) was measured using the AnterUnitherm™ Model 2022 apparatus, which uses the guarded heat flow meter method, at 25 °C in accordance with ASTM standard E1530-11 (2011). The sound absorption coefficient of a material is defined as the ratio of the sound energy absorbed to the total energy impact. The sound absorption coefficient was determined according to the impedance tube method following ISO standard 10534-1 (1996). The equipment consisted of a tube with the sample fixed at one end and a speaker at the other. A microphone was attached to a moving carriage. Sets of samples for the cylindrical products were tested at four selected frequencies: 125, 250, 500, and 1,000 Hz.

In the case of solid bricks, samples were tested at frequencies of 1, 2, and 4 kHz. The sound absorption behavior of the cylindrical gypsum-based products was tested only for the samples with fine wood and rubber particles at the lower 25% proportion (sample codes GR_{f25} and GW_{f25}), which had similar compressive strengths. Finally, the content of volatile organic compounds (VOCs) was measured on a 0.25 m² surface manufactured from the material used in the solid bricks, according to ISO standard 16000-9 (2006). The volume of the room was 0.25 m³, the rate of air exchange was 0.25 m³/h, the load factor was 1 m²/m³, the temperature was 23.1 °C, the relative humidity was 54.3%, and the duration of the test was 168 h.

RESULTS AND DISCUSSION

Gypsum-based Cylindrical Products

Results of compression testing revealed a significant reduction of the ultimate strength of all gypsum-rubber and gypsum-wood cylindrical products as compared to that of the pure gypsum samples (Table 3).

It should be noted that the densities of sample couples GR_{c25}-GR_{f25}, GR_{c50}-GR_{f50}, GW_{c25}-GW_{f25}, and GW_{c50}-GW_{f50} were highly different from each other, despite the same weight ratios of the sample couples. The reason is quite obvious because, for the same material (wood or rubber), when the particle size is reduced (from coarse to fine) the bulk density also decreases. For the same weight, the volume of fine particles is greater than the coarse ones, and therefore the bulk density is smaller.

Table 3. Compressive Strength of Gypsum-based Cylindrical Products (mean values \pm standard deviations)

Product / Product Code *	Density (kg/m ³) **	Compressive Strength (MPa)	Mean Reduction in Compressive Strength (%)
Gypsum			
G	870 \pm 44	2.51 \pm 0.83	-
Gypsum-rubber (GR)			
GR _{c25}	912 \pm 4	0.55 \pm 0.06	78
GR _{c50}	975 \pm 13	0.34 \pm 0.02	86
GR _{f25}	847 \pm 14	0.47 \pm 0.03	81
GR _{f50}	634 \pm 23	0.17 \pm 0.02	93
Gypsum-wood (GW)			
GW _{c25}	779 \pm 44	1.22 \pm 0.12	51
GW _{c50}	441 \pm 19	0.20 \pm 0.13	92
GW _{f25}	656 \pm 8	0.47 \pm 0.07	81
GW _{f50}	417 \pm 48	0.06 \pm 0.01	98

* see Table 1 for labeling
** determined by weighing and calculating the volume of cylindrical samples

After adding wood particles, the products became lighter (417 to 779 kg/m³) than the pure gypsum bricks (870 kg/m³) but the loss in compression strength was significant (51 to 98%). The results are consistent with the strength losses observed in similar concrete-based composites when the waste fractions (*e.g.* cotton, wood sawdust) are increased in the mixture (Turgut 2007; Algin and Turgut 2007). Comparably high strength losses, ranging between 78 and 93%, were noted for the heavier (634 to 975 kg/m³) gypsum-rubber products. Obviously, reinforcement of the compressive strength of the products is needed and could be obtained using additional fibrous materials (virgin or from wastes). As a result of their higher density, the coarser rubber and wood fractions yielded better results than the fine fractions. Increasing the wood and rubber proportions from 25 to 50% reduced strength in both the coarse and fine fractions, especially for wood (Fig. 2).

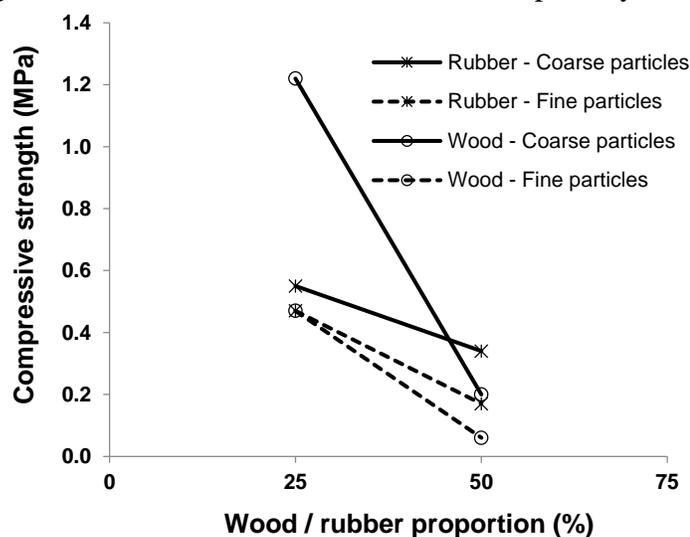
**Fig. 2.** Decrease of compressive strength of gypsum-based cylindrical products with increasing wood and rubber proportions from 25 to 50% for both fractions (fine, coarse particles)

Table 4. Thermal Conductivity Coefficient (k) of Gypsum-based Cylindrical Products (mean values \pm standard deviations)

Product / Product Code *	Thermal Conductivity Coefficient, k (W/m K)
Gypsum	
G	0.314 \pm 0.003
Gypsum-rubber (GR)	
GR _{c25}	0.322 \pm 0.002
GR _{c50}	0.310 \pm 0.002
GR _{f25}	0.291 \pm 0.002
GR _{f50}	0.266 \pm 0.003
Gypsum-wood (GW)	
GW _{c25}	0.312 \pm 0.004
GW _{c50}	-
GW _{f25}	0.262 \pm 0.003
GW _{f50}	0.209 \pm 0.002

* see Table 1 for labeling

Thermal conductivity coefficients ranged between 0.266 and 0.322 W/m K for the gypsum-rubber products and between 0.209 and 0.312 W/m K for gypsum-wood products (Table 4). These values did not differ substantially from those of the gypsum controls (0.314 W/m K) and were also comparable with the range of thermal conductivity coefficients (0.189 to 0.486 W/m K) reported for wood-gypsum (0 to 35%) board (Bekhta and Dobrovoska 2006). The cylindrical products provided better thermal insulation than concrete (1.396 W/m K) (Xu *et al.* 2004) and pine wood (0.450 to 0.630 W/m K). Plywood and particleboard provide better thermal insulation, with values of 0.083 and 0.097 to 0.133 W/m K, respectively (Xu *et al.* 2004; Nemli and Colacoglou 2005). Improved thermal insulation was observed with fine rubber and wood fractions (Table 4). It should be mentioned that wood's thermal conductivity depends on a number of factors such as the species, density, moisture content, and temperature (Tsoumis 1991). In composites, thermal conductivity depends on the particle size of the constituent materials and on density. In particleboards, density is positively correlated with thermal conductivity (Khedari *et al.* 2003).

Table 5. Sound Absorption Coefficients of Gypsum-based Cylindrical Products and Wood-based Products at Frequencies from 125 to 1,000 Hz

Product	Density (kg/m ³)	Thickness (mm)	Sound Absorption Coefficient
Gypsum-rubber (GR _{f25}) *	847	50	0.18 to 0.36 **
Gypsum-wood (GW _{f25}) *	656	50	0.19 to 0.47 **
Rubber Granulated Panel	-	-	0.60 to 0.70 ***
Wood Board (pine)	520	19	0.09 to 0.12 ***
Plywood	550	12	0.04 to 0.25 ***
Particleboard	-	20	0.06 to 0.26 ***
Low-density Particleboard	300	30	0.06 to 0.65 ***
Insulation Fibreboard	220	13	0.04 to 0.69 ***
Low-density Fibreboard	200	12	0.06 to 0.71 ***

* see Table 1 for labeling; ** this study; *** taken from Xu *et al.* 2004

Products with wood had slightly better sound insulating capacity as compared to those with rubber (Table 5). This very small difference should be attributed to the lower density of the gypsum-wood product and also to the fact that air-dry wood has a better sound-insulating capacity than rubber. The gypsum-based wood and rubber products had a higher sound absorption coefficient than wood, plywood, or particleboard. However, their ability to absorb sound is much lower than that of traditional insulating products such as insulating board (Table 5). Sound absorption could be improved by leaving void spaces (holes) inside the products. It is known that sound-proofing as well as thermal properties of gypsum building materials can be improved by increasing the porosity (Vimmrova *et al.* 2011).

Gypsum-based Solid Bricks

The density of the gypsum-based solid bricks (Fig. 3) was $580 \pm 23 \text{ kg/m}^3$. Taking into account that every brick has a known void volume (the holes represent 22.76% of the brick's volume) of 196.05 cm^3 , the density of the material used for brick manufacturing was $750 \pm 30 \text{ Kg/m}^3$.



Fig. 3. Gypsum-based solid bricks with six symmetric round holes measuring $85 \times 55 \times 185 \text{ mm}^3$ ($W \times T \times L$) and density of $580 \pm 23 \text{ Kg/m}^3$

In terms of compressive strength, the bricks performed slightly better when loaded at the large surface of the lateral upper side than at the small surface. In the first case, the ultimate stress in compression was $0.57 \pm 0.03 \text{ N/mm}^2$ and $0.50 \pm 0.01 \text{ N/mm}^2$ in the second. For both large and small surface loading, failures occurred toward the direction of the load in the material under a hole or between the rows of holes. The strength in compression was almost double in the axial direction, with a mean value of $1.09 \pm 0.20 \text{ N/mm}^2$. Failures of bricks in axial compression were diverse. The results of the compressive strength testing of bricks were used for static calculations needed in internal wall construction. Two different construction techniques were considered.

In the first technique, a 3 m-high internal wall was envisioned with the bricks placed one on top of another with their small surfaces ($55 \times 185 \text{ mm}^2$) in contact. In this configuration, a 3 m-high wall would require 35 bricks stacked in a single column from the floor to the top of the wall. According to the compression testing results, each brick can withstand around 5088 N, 518 kg, or 0.50 N/mm^2 , much greater than the weight (approximately 20 Kg) or load (approximately 196 N or 0.02 N/mm^2) of a stack of 35 bricks.

In the second technique, 3 m-high internal wall was envisioned with the bricks placed one on top of another with their large surfaces ($85 \times 185 \text{ mm}^2$) in contact. In this

case, a 3 m-high wall would require 55 bricks stacked in a single column from the floor to the top of the wall. According to the compression testing results, each brick can withstand around 8932 N, 911 kg, or 0.57 N/mm², much greater than the weight (approximately 31 kg) or the load (approximately 304 N or 0.02 N/mm²) of a stack of 55 bricks.

The thermal conductivity coefficient (k) of solid bricks was calculated as 0.274 W/m K. The bricks exhibited better thermal insulation than either the extruded or pressed house bricks, which according to the literature have thermal conductivity coefficients of 0.33 to 0.98 and 0.87 to 1.10 W/m K, respectively. However, the insulating bricks perform much better as they have a thermal conductivity coefficient of 0.15 W/m K (Ramachandran *et al.* 2002).

Table 6. Volatile Organic Compounds (VOC) Emissions of Gypsum-based Bricks

Test	Duration (168 h)
Formaldehyde Emissions (mg/m ³)	0.012
Acetaldehyde Emissions (mg/m ³)	0.083
Toluene Emissions (mg/m ³)	0.022
Tetrachlorethylene Emissions (mg/m ³)	0.0027
Xylene Emissions (mg/m ³)	0.069
1,2,4-Trimethylbenzene Emissions (mg/m ³)	<0.0001
Dichlorobenzene Emissions (mg/m ³)	<0.0001
Ethylbenzene Emissions (mg/m ³)	<0.0001
Butoxyethanol Emissions (mg/m ³)	<0.0001
Styrene Emissions (mg/m ³)	0.0059
Total VOCs Emissions (mg/m ³)	0.051

The maximum sound absorption coefficient of the solid bricks was set to the frequency of 1 kHz (0.72) and decreased to equal levels of 0.43 and 0.45 with increases in the frequency from 2 to 4 kHz, respectively. Measurements at low frequencies should provide more information on the acoustic behavior of bricks across the entire frequency range and would make a comparison with other materials behavior, for which available data exist only for the low frequency range of 125 to 1,000 Hz, possible. It should be noted that before acoustic testing, the bricks were also tested for their air flow resistance. Very high values were obtained as compared with that of a wool commercial rock (URSA) of 2.5 cm thickness, implying that the resistance to air flow is high and that the bricks have a very low degree of porosity. Thus, it was not possible to test the present material at low frequencies.

The results presented in Table 6 showed low VOCs emissions from the bricks, which correspond to “class A” emissions according to the labelling required by the relevant French law for construction products, covering walls, floors, and coatings (French Republic 2011). The emissions level of the product is defined by a class ranging from A+ (very low emissions) to C (high emissions), corresponding to the principle already used for electrical appliances and vehicles (whole class: A+, A, B, C).

CONCLUSIONS

1. From a mechanical point of view, coarse fractions of wood and rubber are preferable for use in the manufacture of gypsum-based products. Also, the proportion of the wood or rubber particles added should not exceed 25% of the total weight. Mechanical reinforcement of the products could be acquired using various fibrous materials.
2. Thermal conductivity was similar in all wood- and rubber-type of products. It was also independent of the material (gypsum, rubber, wood), particle size, and material proportions.
3. For sound absorption, no substantial differences were observed between the gypsum-rubber and gypsum-wood products.
4. The overall properties of the gypsum-based solid bricks were promising for further investigation of the manufacturing and properties of wall materials based on recovered wood and rubber. The bricks provide new usage opportunities for the rubber-textile material, a waste generated in the tire recovery process and presently without any other use.
5. In terms of compressive strength, the bricks performed slightly better when loaded at the large surface of their lateral upper side than at the small surface. Furthermore, a single brick can bear the load of 35 or 55 bricks, as required for a single column when 3 m-high walls are constructed with bricks in contact at their small or large surfaces, respectively.
6. Thermal insulation and VOC emissions of bricks were acceptable, but experimentation is needed regarding sound absorption.

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