Influence of expanded vermiculite on physical properties and thermal conductivity of clay bricks

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Abstract

Porous clay bricks lightened by adding 2.5, 5, 7.5 and 10 wt.% expanded vermiculite have been fabricated by semi-dry pressing process. The expanded vermiculite (in Yildizeli, Sivas, Turkey) was used as an additive into a brick raw material to produce the porosity. Chemical composition, phase identification, thermal behavior and microstructure of the raw materials were analyzed by XRF, XRD, TGA and SEM, respectively. The brick mixtures containing vermiculite at different proportions were formed, dried and then fired at 900 and 1000 °C for two hours. Properties such as drying and firing shrinkages, loss on ignition, bulk density, porosity, water absorption, compressive strength, thermal conductivity and microstructure of the samples were determined. It is found that the use of expanded vermiculite addition reduced the bulk density of the samples containing 10 wt.% additive down from 1.76 to 1.34 g/cm³. It was observed that their porosity ratios up to 45% improved with increasing of vermiculite addition, whereas their compressive strengths (min. 14 MPa) decreased. However, their strengths were still quite higher than that of required by the standards. Thermal conductivity of the porous samples with vermiculite of 10% decreased from 0.96 to 0.65 W/mK by raising porosity, which corresponds to a reduction of 32% compared to the reference sample. Increasing of the firing temperature also affected their mechanical and physical properties. In consequence, this study revealed that the brick samples produced with vermiculite addition could be used as an insulating material in construction applications.

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Keywords: Lightweight bricks; Expanded vermiculite; Pore-forming; Thermal insulation; Physical properties

1. Introduction

Energy savings is recently one of the most important issues in the world because of both economic and environmental concerns. Nowadays, almost one third of the overall energy consumed in the world is eventually used in buildings for heating and cooling [1–3]. Many countries aim to increase the energy efficiency of buildings. An important way of achieving better energy efficiency in buildings is to improve their thermal insulation properties. This can be done by either micro-pores generated by pore-maker additives before bricks were fired, or by introducing holes extending through the brick like in the case of a perforated brick [4,5]. Thus, there are two different thermal conductivity values of construction brick materials: first involves the bulk of the material constituting the walls, while the second involves equivalent thermal conductivity of the entire product consisting of large vertical perforations of rectangular cross-section [4,6]. In the former, when pore-making additives are used in clay brick, they burn and occurred micro-pores which help reduce thermal conductivity of the brick [7–9]. The latter is used widely by the brick industry to save clay material, to reduce the weight of the product and also to decrease thermal conductivity of the brick. The thermal performance of bricks depends on the geometry of the brick recesses and the material properties. The differences of thermal conductivity in function of product geometry are described another study by Sutcu et al. [10].

In general, thermal conductivity of the bulk clay bricks is approximately 1.0 ± 0.4 W/mK depending on their raw materials, processing, firing temperatures and fired densities [4,11–13]. These values can be reduced by addition of various organic and inorganic pore-makers into the brick raw material mixtures before firing.
When this is made into an extruded product with vertical perforations its thermal conductivity could be much lower as 0.08 W/mK \[5,6\]. Different pore-forming materials that act by thermal decomposition and volatilization (e.g. wood sawdust, polystyrene, organic residues, coal dust, powder limestone, papermaking sludge) and heat-resistant porous materials (e.g. pumice, diatomite and perlite) in brick body have been widely used \[7–9,14–18\]. Yet, very limited information on the usage of vermiculite as pore maker in brick and ceramic production has been reported \[4,19\]. In this study, expanded vermiculite in milled powder form as an inorganic pore-making additive is used for production of porous and lightweight clay bricks.

In the world, vermiculite ores is mostly found in South Africa (\(\sim 41\%\)), the USA (\(\sim 21\%\)) and China (\(\sim 21\%\)) \[20\]. In Turkey, totally 5.2 Mt reserve of vermiculite is reported by Mineral Research & Exploration General Directorate. The studies performed in recent years indicated that the most important deposits of Turkey are Karakoç mines (in Yıldızeli, Turkey) where are more economic than the others due to the higher of their expansion ratios \[21\]. In the literature, the term of “vermiculite” is used to describe commercially exploited deposits of micaceous minerals (variable mixtures of different minerals like vermiculite, hydrobiotite and phlogopite) which can be exfoliated when heated rapidly to high temperatures \[22\]. Vermiculite mineral has a mica-like lamellar structure that quickly expands on heating to produce a lightweight material. Vermiculite is formed by weathering or hydrothermal alteration of hydrobiotite or phlogopite (KMg\(_3\) Al\(_2\)Si\(_4\)O\(_{10}\)(F,OH)\(_2\)) mineral phases \[22,23\]. When heated rapidly, vermiculite exfoliates as the interlayer water turns into steam, forcing the silicate layers apart from one another in an accordion-like expansion with expansion ratio of 20–30 times its original thickness depend on the temperature and time of processing \[24–26\]. The thermal expansion characteristics of Karakoc (in Turkey) deposits were determined in flame and electric furnace conditions and maximum expansion ratios were reported as 13 and 18 times, respectively \[25\]. In addition, its chemical exfoliation characteristics showed higher exfoliation rates. According to this study, these deposits included mostly vermiculite and phlogopite phases due to the alteration of micas \[27,28\].

Expanded vermiculite has very low density, good thermal and acoustic insulation properties, and also is a chemically inert and fire resistant material, which makes it attractive for use as lightweight aggregate and filler for heat insulation applications \[29–33\]. The aim of this study was to determine of feasibility of using the expanded vermiculite powder in production of clay brick samples and the effects on physical and mechanical properties and thermal conductivity of bricks.

### 2. Materials and method

In this study, production of brick samples from vermiculite added clay mixtures were accomplished with the purpose of using as an insulating construction material. The effect of expanded vermiculite additive of 2.5%, 5%, 7.5% and 10% by weight in finely powder form on the physical and thermal properties of clay bricks is investigated. The clay raw material was obtained from a brick manufacturer (in Bartın, Turkey). The vermiculite was taken from Demircilik vermiculite quarry (in Yıldızeli, Sivas, Turkey). Expanded vermiculite is obtained by heating raw vermiculite at 600 °C for 10 s. The brick raw material and expanded vermiculite was initially subjected to pretreatments such as drying, milling and sieving. In this study, the raw materials with particle size smaller than 150 μm was used for brick production. The physical properties of the vermiculite are presented in Table 1.

The raw materials were initially characterized by chemical, mineralogical, thermal and microstructural analysis. The chemical composition of the raw materials was determined by the X-ray fluorescence (Spectro IQ II XRF) spectrometer. Their mineralogical phase content was identified by using a Phillips X’Pert Pro X-ray powder diffractometer (Cu-Kα radiation, \(\lambda = 1.54056 \text{ Å}, 40 \text{ mA}, 40 \text{ kV}\)). The thermo-gravimetric and differential thermal analyses of the raw materials were performed at a heating rate of 10 °C/min in nitrogen atmosphere using the thermal analyzer instrument (Perkin Elmer Diamond TG/DTA). Also, their microstructural images and elemental maps were investigated by a scanning electron microscopy (FEI Quanta250 FEG SEM) analysis with energy dispersive X-ray spectrometer (EDS).

In this study, experimental flowchart for brick production process is shown in Fig. 1. Expanded vermiculite in powder form was added into the brick clay raw material at ratios of

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**Table 1**

Properties of expanded vermiculite (The data provided by the manufacturer).

<table>
<thead>
<tr>
<th>Characteristic</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Color</td>
<td>Silver</td>
</tr>
<tr>
<td>Shape</td>
<td>Accordion shaped granule</td>
</tr>
<tr>
<td>Water holding capacity</td>
<td>240 wt%</td>
</tr>
<tr>
<td>Cation exchange capacity</td>
<td>90 meq/100 g</td>
</tr>
<tr>
<td>pH (in water)</td>
<td>6.1</td>
</tr>
<tr>
<td>Thermal conductivity value</td>
<td>0.063 W/mK</td>
</tr>
<tr>
<td>Combustibility</td>
<td>Non-combustible</td>
</tr>
<tr>
<td>Sintering temperature</td>
<td>1170 °C</td>
</tr>
<tr>
<td>Specific heat</td>
<td>0.22 kcal/kgK</td>
</tr>
<tr>
<td>Bulk density</td>
<td>140 kg/m(^3)</td>
</tr>
</tbody>
</table>

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**Fig. 1.** Experimental flowchart for brick process.
2.5, 5, 7.5 and 10 wt.%. The mixtures were mechanically blended with about 18 wt.% of total weight sprayed water for 30 min in a laboratory mixer to the desired homogeneity and consistency depending on the amount of additive. The blends were poured into a plaster bowl to reduce the amount of moisture. Mixtures were allowed to stand in the bowl in order to suck of water by capillary effect for one hour. The moisture-reduced blends had a proper consistency to form the brick samples. Three rectangular brick samples (20 × 60 × 100 mm) for each test, from proper consistency mixtures, were semi-dry pressed into a steel mold with a hydraulic press under a pressure of 10 MPa, then the samples that carefully removed from the mold were kept waiting in ambient conditions for overnight. The samples were firstly dried at 40 °C for 20 h and then at 100 °C for 18 h in an oven. After drying, the samples were fired in a laboratory-type electrical furnace (Protherm PLF12/15) at the rate of 1 °C/min until 600 °C and then at the rate of 5 °C/min until the dwell temperatures of 900 °C and 1000 °C for 2 h to achieve strength.

The fired samples were characterized for the physical, mechanical, thermal conductivity and microstructural properties. The bulk density, apparent porosity and water absorption values were measured with Archimedes method applied to the samples. Their microstructures and compressive strengths were determined. Thermal conductivities of the brick samples were performed by the C-Therm TCI Thermal Conductivity Analyzer with modified transient plane source at ambient conditions.

3. Results and discussion

3.1. Characterization of the raw materials

The chemical composition measured by XRF of the raw materials is given in Table 2 in oxide form. According to this, the brick clay contains a large fraction of silica as well as the oxides of aluminum, iron, calcium, magnesium and potassium. The vermiculite includes mainly the oxides of silicon, aluminum, magnesium and iron. The loss on ignition (LOI) of brick clay and vermiculite on heating at 1000 °C was measured as percentages of 8.6 and 9.2, respectively.

The XRD patterns of raw materials show the mineral phases in Fig. 2. According to the XRD analysis, the brick clay includes mainly quartz, muscovite, illite and fewer amounts of chlorite constituents (Fig. 2a). Fig. 2b shows XRD patterns of the expanded vermiculite before and after grinding. The expanded vermiculite consists mainly of vermiculite, phlogopite mica and hydrobiotites, likely an interstratified. According to X-ray patterns, it was observed that the crystallinity of vermiculite changed after the milling. Interstratified structure of vermiculite after milling does not contain a hydrated phase, vermiculite (two layers of water molecules in interlayer) and the mica-type phase (zero water molecules in the interlayer) still remain.

The thermal gravimetric (TG) analysis of the brick clay and vermiculite are presented in Fig. 3a and b, respectively. In the TG curve of the brick clay (Fig. 3a), its weight loss for 1000 °C was about 7%, the first 1% decrease in the mass occurred about 100 °C due to the evaporation of physical water. The second (1%) and third (2%) weight losses were observed between the 200–300 °C and the 350–550 °C ranges which may be due to the burning of organic matter and the removal of chemical water, respectively.

Table 2
Chemical composition of the raw materials used (% by weight).

<table>
<thead>
<tr>
<th>Oxides</th>
<th>Brick clay</th>
<th>Expanded vermiculite</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO₂</td>
<td>59.2</td>
<td>36.9</td>
</tr>
<tr>
<td>TiO₂</td>
<td>0.95</td>
<td>2.18</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>14.9</td>
<td>17.7</td>
</tr>
<tr>
<td>Fe₂O₃</td>
<td>8.54</td>
<td>11.2</td>
</tr>
<tr>
<td>MgO</td>
<td>1.64</td>
<td>16.4</td>
</tr>
<tr>
<td>CaO</td>
<td>2.36</td>
<td>3.54</td>
</tr>
<tr>
<td>Na₂O</td>
<td>0.96</td>
<td>0.15</td>
</tr>
<tr>
<td>K₂O</td>
<td>2.86</td>
<td>2.64</td>
</tr>
<tr>
<td>Loss on ignition</td>
<td>8.6</td>
<td>9.2</td>
</tr>
</tbody>
</table>

Fig. 2. X-ray diffraction patterns of (a) the brick clay and (b) expanded vermiculite (EV) and milled EV.
The largest weight loss (2.5%) was detected in between the 550–800 °C range which is possibly related with the dehydroxylation of illite and chlorites [4]. The TG curve of expanded vermiculite is given in the Fig. 3b where a total weight loss of about 10% for 1000 °C. The weight losses are related to the adsorbed and interlayer water. In the first weight loss, the dehydration of the vermiculite takes place between 20 and 110 °C which corresponds to the loss of free water physically absorbed on the surface. The mass losses in the range of 100–200 °C can be attributed to the removal of more free water molecules residing in the particle

![TGA curves](image)

Fig. 3. TGA curves of the raw materials: (a) brick clay and (b) expanded vermiculite.

![SEM images](image)

Fig. 4. SEM images of (a, b) expanded vermiculite, (c) vermiculite powder after milling and (d) the brick clay at different magnifications.
spaces of the vermiculite [34]. The mass losses in the temperature range of approximately 500–800 °C can be attributed to the loss of interlayer water and chemical water molecules, which are in contact with the cations in the interlayer region [19]. The region between 800 °C and 1000 °C records endothermic peaks, which could be assigned to the formation of new crystalline phases [34].

Fig. 4 shows the scanning electron micrographs of raw materials used in this study. In Fig. 4a and b, the SEM images of an expanded vermiculite grain are shown at different magnifications. The size of expanded vermiculite is smaller than 5 mm and the vermiculite flakes that expanded in an elongated accordion-like form are shown in Fig. 4a. In Fig. 4b, it is observed that has the silicate layers separated from each other as a result of rapidly heating. Fig. 4c indicates the milled vermiculite flakes with a size range of smaller than 150 μm. The SEM morphology of the clay raw materials revealed that it consisted of plate-like particles as well as agglomerated particles with sizes that smaller than 20 μm (Fig. 4d). According to the EDS analysis, the vermiculite includes 42.6 ± 2.8% SiO₂, 18.5 ± 1.1% Al₂O₃, 15.6 ± 1.1% Fe₂O₃, 13.1 ± 2.8% MgO, 3.4 ± 0.2% CaO, 2.6 ± 0.3% TiO₂ and 2.6 ± 0.1% K₂O in oxide form. These results showed to be close to the XRF analysis.

3.2. Characterization of the fired samples

Figs. 5–7 show the experimental results obtained from the brick samples fired at 900 °C and 1000 °C, where the term of V indicates the amount of vermiculite added into the mixtures. Their drying shrinkage values changed until max. 2%. The brick samples showed the dimensional stability during firing and their linear firing shrinkages were almost smaller than 1%.

The bulk density, apparent porosity and water absorption values of the produced samples for different proportions of expanded vermiculite fired at different temperatures were presented in Fig. 5. As shown in Fig. 5, the bulk density of the brick samples without addition (indicated as V0) fired at 900 °C was 1.76 g/cm³, in addition, it had a porosity of 31.3% and water absorption of 17.8%. For this brick, the compressive strength of 31.7 MPa was quite well. The bulk density of the brick samples decreased from 1.76 g/cm³ to 1.34 g/cm³ as the amount of expanded vermiculite increased from 0% to 10%. That may due to the expanded vermiculite has relatively low unit weight, and the vermiculite particles are layered and expanded, which led to a reduction in the bulk density of the clay bricks. Depending on the increase in the vermiculite addition, the apparent porosities and water absorption

![Graphs showing bulk density, apparent density, water absorption, and apparent porosity results of the samples.](image-url)
the noticed all the samples show an increasing bulk densities with turing parameters affecting the brick properties. It can be enough to produce these type-bricks. produced bricks will be useless. For this study, the ratios used it is water absorption ratios will excessively increase. In this case, 30% are generally the maximum values acceptable[36]. If higher porosity of bricks. For clay bricks, the water absorption of up to durability of bricks[35]. Therefore, it was closely related to the clay bricks. When water absorption values increase, it decreases the water absorption is an important factor affecting the durability of bricks. That may due to higher temperature significantly depending on the vermiculite addition and clay bricks. When water absorption values increase, it decreases the durability of bricks [35]. Therefore, it was closely related to the porosity of bricks. For clay bricks, the water absorption of up to 30% are generally the maximum values acceptable [36]. If higher rates of vermiculite additive are added into the brick mixtures, the water absorption ratios will excessively increase. In this case, produced bricks will be useless. For this study, the ratios used it is enough to produce these type-bricks.

Also, effect of the firing temperature was investigated on the samples. The firing temperature is one of the main manufacturing parameters affecting the brick properties. It can be noticed all the samples show an increasing bulk densities with the firing temperature rising, while a decreasing porosity and water absorption values. That may due to higher temperature yielded the deformation of vermiculite flakes.

In Fig. 6, the compressive strength of the samples varied significantly depending on the vermiculite addition and firing temperatures. Accordingly, the strength of the samples fired at 900 °C decreased from 31.7 to 14.2 MPa with increasing of additive amounts, but the lowest strength value of the sample is quite higher than the Turkish and corresponding European Standard minimum value of 7 MPa [35]. This is related with the porosity or density of the fired solid bricks [4]. In addition, increasing of firing temperature partially increased their strengths.

The thermal conductivity test results of the brick samples are presented in Fig. 7. Their thermal conductivities considerably decreased with increasing of vermiculite addition. Thermal conductivities of the bricks depend on their firing temperatures, densities and therefore porosities of the samples. The thermal conductivity values also significantly increased with rising of the firing temperature. It can be clearly seen that the thermal conductivity of brick samples is closely related with their porosities or densities and also firing temperatures. Thermal conductivity of the porous sample with 10% vermiculite addition (V10 sample) fired at 900 °C (0.65 W/mK) showed more than 30% reduction compared to the reference brick (0.96 W/mK). The thermal conductivity in term of physical properties of the bricks relatively lowered more than the bulk density drop. Efficiency of the expanded vermiculite on the physical properties is clearly seen from the results. This can be attributed to the total porosity, the size and the nature of the pores between the vermiculite flakes. The reduction in thermal conductivity of the brick samples produced in this study is quite encouraging for higher energy saving potential in building applications. If the bricks are produced as a perforated brick from these compositions, the equivalent thermal conductivity will be reduced to more satisfactory values.

The general SEM images in backscatter electron (BSE) mode of the V10 brick samples fired 900 and 1000 °C are shown in Fig. 8, respectively. The micrographs of fired clay bricks with additives revealed the porous structure which is helpful in reducing their fired densities. Coarse particles with dimensions of 100 μm identified as quartz by EDS analysis are observed in the structure. Some large pores that existed in the body could be a pull-out during polishing. The vermiculite flakes in layered structure are also observed in the microstructure of samples. These flakes generated the large amounts of porosities in the brick body. The EDS analysis of the layered structures in Fig. 8d consisted of 42.2 ± 2.4% SiO2, 16.9 ± 1.4% Al2O3, 3.0 ± 0.5% TiO2, 16.6 ± 1.3% Fe2O3, 1.1 ± 0.2% K2O, 4.0 ± 0.3% CaO, 15.9 ± 2.1% MgO and 0.3 ± 0.1% Na2O, approximately. The brick matrix also occurred of about 60.9 ± 1.2% SiO2, 18.0 ± 0.8% Al2O3, 1.7 ± 0.3% TiO2, 7.5 ± 0.4% Fe2O3, 2.4 ± 0.3% K2O, 7.1 ± 0.2% CaO, 1.7 ± 0.3% MgO and 0.5 ± 0.1% Na2O.

Increasing of the temperature caused their porosity ratios minor decreased. It is possible that due to the beginning of vitrification in the brick structure, and if the firing temperature is increased the degree of densification rises and in the event, reduces the connection between the pores in the flakes [14]. In this study, the selected working temperature of 900 °C is ideal for available brick features.

The XRD analysis of fired bricks with vermiculite addition was performed for the crystalline phases occurred in the brick bodies. Fig. 9a and b shows the XRD patterns of fired brick samples without vermiculite and the sample with 10wt.% vermiculite addition, respectively. The results obviously indicated that fired
reference brick samples without vermiculite additive consist of quartz (SiO₂) on a large scale, and also hematite (Fe₂O₃) and sanidine (KAlSi₃O₈). Vermiculite containing-bricks consist mainly of quartz, hematite and sanidine as well as phlogopite. As addition of vermiculite increased, amount of phlogopite phase increased. However, crystalline phlogopite peaks decreased with increasing of firing temperature of the bricks. This shows a morphological change occurred in vermiculite flakes under firing of 1000 °C. Probably, phlogopite mica improved vitrification degree in brick clay body [37].

4. Conclusions

In this study, the usable of vermiculite from natural raw material sources of Turkey in the brick production was investigated. Clay brick samples containing expanded vermiculite powder with different percentages were prepared by using of vermiculite raw material mined from Turkey. Properties of the brick samples produced at different firing temperatures are presented below.

- The bulk density of bricks decreased with increasing vermiculite additive, while their porosity and water absorption values increased.
- All samples have sufficient strength values greater than 14 MPa.
- Thermal conductivity of the microporous brick sample with 10% vermiculite produced at 900 °C compared to the brick without additive, decreased from 0.96 to 0.65 W/mK.
- Effect of the firing temperature on the properties of the bricks was additionally investigated. Increasing of the firing temperature affected the properties of the bricks, which increased their mechanical strength and thermal conductivity, besides decreased their porosities.
The results showed that the brick samples produced with vermiculite addition could be used as a construction material for insulating applications. Also, if a perforated clay brick from the best composition in this study is produced, the equivalent thermal conductivity of the wall which made from these bricks will be reduced to more satisfactory values.

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References


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