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Reducing CO_2 Emissions in the Production of Porous Fired Clay Bricks

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A plan to reduce CO_2 emissions is a priority these days. Brick industry contributes to the increase of these emissions mainly through the use of combustible pore-forming agents such as sawdust, cellulose, and coal sludge. These agents are used to improve the thermal insulation properties of brick products, and the suppliers regularly increase the prices of these agents based on their high consumption. Therefore, in an effort to reduce raw material expenses and CO_2 emissions, brick manufacturers are looking for new possibilities while maintaining the quality of their products. This article discusses the possibility of using industrially manufactured product Vuppor as an additive as a replacement for combustible pore-forming agents. The presence of this additive in the fired clay body increases the proportion of pores, especially with a size range between 0.1 and 5 µm, having a positive impact on the reduction of its thermal conductivity. With a 0.5 wt.% dose of Vuppor additive, the brick production costs and thermal conductivity can be reduced by 20 % and 12 %, respectively, while also achieving reductions in CO_2 emissions over 60 %. Consequently, the combustible pore-forming agents can be used in a more environmentally friendly manner, for example in the furniture industry, the biogas production, and the like.

Keywords: brick, pore-forming agent, thermal conductivity, pore structure, production cost.

1. INTRODUCTION

Nowadays, brick industry is characterized by the use of high amounts of combustible pore-forming agents like sawdust and cellulose and coal sludge [1 – 3]. This trend was caused by customers who prefer products with high thermal insulation properties. Some of the factors that have been monitored closely with regard to the addition of combustible pore-forming agents are:

- use as raw material in the production of bricks [4 – 9];
- reduction in the amount of fossil fuels used in the process of firing [10, 11];
- reduction in thermal conductivity by increasing the porosity of the fired clay body [12 – 15];
- reduction in thermal conductivity by regulating pore structure of the fired clay body [16 – 18].

Unfortunately, in recent years, construction firms have put brick products manufacturers under pressure in order to achieve products with a very high thermal resistance value. This resulted in an overuse of combustible pore-forming agents, which ultimately caused the formation of the so-called "black core", see Fig. 1.

The black core may be incurred at brick products whose base raw material contains a high proportion of organic combustible material and also contains a relatively high proportion of carbonates. Then at a fast growth of the firing temperature leads to compaction of the surface layer of fired clay body which causes slowing diffusion of oxygen between the inside and the outside. Lack of oxygen inside the fired clay body then causes the formation of charcoal – black core [19].

Fig. 1. Detail of the black core in fired clay body

Nowadays, as a response to the high demand, suppliers of combustible pore-forming agents regularly increase their prices. Thus, brick manufacturers, in an effort to reduce raw material expenses and CO_2 emissions while maintaining the quality of their products, are looking for new solutions. One such solution could be the addition of Vuppor. By applying Vuppor additive to the clay raw material, their properties and some of the related technological processes can be improved [20, 21]:

- shortening of the drying process due to a decrease in its drying sensitivity value, which will improve the quality of the dried products without cracking;
- shortening of the burning process and reduction of the firing temperature led by an increase in gas diffusion in the pore system of the fired clay body;
- a reduction in the coefficient of thermal conductivity of the fired clay body, which significantly improves the thermal properties of the brick products, allowing their

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application in the construction of low energy or passive houses.

When using Vuppor, no complications should arise during the production process. All the necessary equipment, like the dosing system, is the same as that used for the production of fresh concrete. In this case, the dosing device should be placed in front of the circular screen feeder.

For the production of bricks, one Slovak manufacturer planned to reduce the associated costs by replacing part of the combustible pore-forming agents with Vuppor additive. By mutual collaboration between the Faculty of Civil Engineering, Slovak University of Technology, Bratislava, and the Faculty of Civil Engineering, Brno University of Technology, we tried to solve this problem through the experiment discussed below.

The solution of this problem was interesting in that the brick clay contained a relatively high proportion of carbonates. From the literature it is known that just a high proportion of carbonates in the brick raw material increases the value of the thermal conductivity of fired clay body and it can also at the high dose combustible pore forming agent to cause problems as indicated above [22].

2. EXPERIMENTAL

For this experiment, we used basic brick clay raw materials consisting mainly of Quaternary loess sediments (loess and loess loams) from Pezinok (Slovak Republic). The chemical and particle size composition of loess sediments are listed in Table 1 and Table 2, and the mineralogical characterization (TGA-DTA and XRD) is shown in Fig. 2 and Fig. 3, respectively.

Table 1. Chemical composition of the brick clay

Chemical composition	%
SiO_2	48.7
Al_2O_3	13.7
Fe_2O_3	5.67
CaO	9.99
MgO	3.74
Na_2O	0.50
K_2O	2.51
MnO	0.35
TiO_2	0,69
P_2O_5	0.16
SO_3	0.21
Loss on ignition	13.4

Pore-forming agents (sawdust 12 vol.%, cellulose sludge 1.5 vol.% and 1 vol.% of coal sludge) are added to the brick clay in order to reduce the thermal conductivity of the final products, Table 3 (sign of testing mixture – M0).

Table 2. Granulometric composition of the brick clay

Grain size	%
under 0.002, mm	22.0
0.002 to 0.2, mm	68.0
above 0.2, mm	10.0

From the mineralogical point of view, the material is composed of illite-chlorite with a high proportion of calcium carbonates there is possible to calculate approximate content of calcite about 15 % according to DTA and TGA curves (Fig. 2) in the temperature range 750 – 850 °C (decomposition of $CaCO_3$).

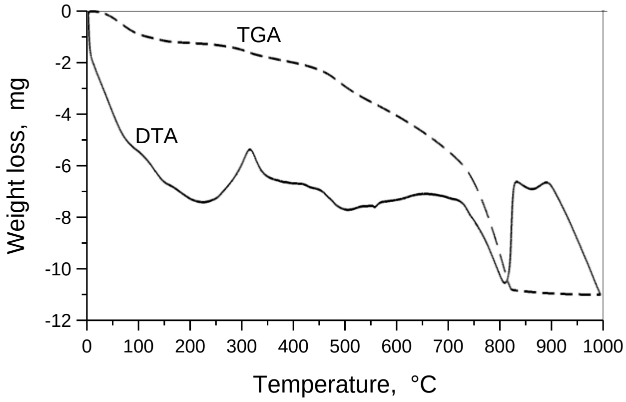

Fig. 2. TGA-DTA curves of the brick clay

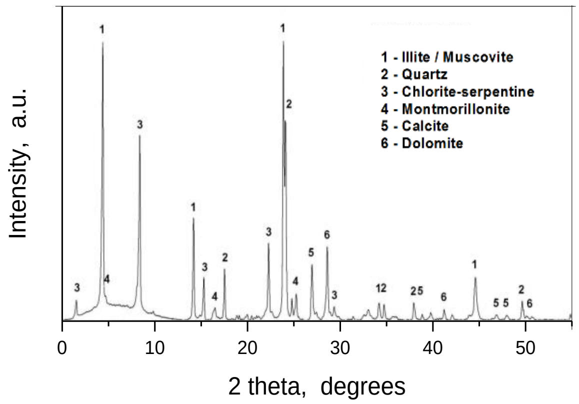

Fig. 3. XRD of the brick clay

Table 3. Test mixtures

Sign of test mixture	Sawdust vol.%	Cellulose sludge vol.%	Coal sludge vol.%	Vuppor wt.%
M0	12	1.5	1	–
M1	12	1.5	1	0.5
M2	12	1.5	1	0.75
M3	–	–	–	–
M4	5	–	–	–
M5	5	–	–	0.5
M6	5	–	–	0.75

Vuppor additive is industrially manufactured product (name company: VUP – Petrochemistry Research Institute, Prievidza, Slovak Republic). This additive is a white emulsion based on aldehyde condensates that contains no sulfur containing compounds or inorganic impurities. During its burning process, only oxides of carbon, nitrogen and water vapor are emitted, its TGA-DTA curves are shown in Fig. 4. Vuppor is organic material with very narrow temperature range of burning about 400 °C. The burning of 1 kg of dry matter produces about 0.502 m^3 of CO_2, this is less than when burning sawdust (0.932 m^3), cellulose (0.875 m^3) and coal sludge (1.307 m^3). Vuppor additive has a dry matter value of 38 wt.% and a density of 1140 kg/m^3, is soluble in water and meets all the hygiene requirements. The optimal dose of the additive, measured in dry matter percentage, ranges from 0.5 to 0.75 wt.% of the total dry clay weight. For this, the total dry matter of the additive is determined gravimetrically by the amount of residue remaining after desiccated.

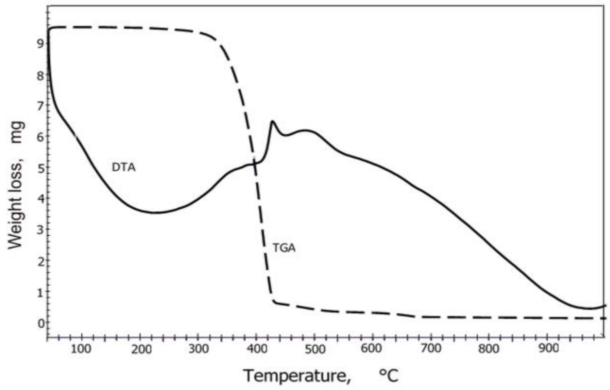

Fig. 4. TGA-DTA curves of the Vuppor additive

The presence of this additive results in a surface tension increase on the water contained in the brick clay, which significantly reduces its plasticity. To achieve the original value of plasticity, the amount of water added must be increased. (Note – The value mass plasticity is different in each factory and depends on the composition of brick raw material and manufacture technology.) This water – working moisture content will evaporate during the drying process, and consequently, higher porosity both on the dried and on the fired clay body can be obtained.

2.1. Production of test samples

According to the requirements of the manufacturer, seven test mixtures were prepared (Table 3). The applied additives (sawdust, cellulose sludge, coal sludge and Vuppor) were added to "clean" clay. Then each mixture was homogenized. The constant mass plasticity was the same in all cases (upsetting height 31 mm according to Pfefferkorn). Plastic clay bodies (sample dimensions of 100 x 50 x 20 mm) were uniaxially pressed at pressure of 1.2 MPa. Test samples were stored for 48 hours at laboratory temperature (20 °C) and then dried at constant weight at 105 °C. Specimens were fired in an electric laboratory furnace with a regulated regime of firing. We have maintained a stable oxidation environment within the furnace and the firing curve applied in the brickworks of Pezinok was used with the maximum temperature of 870 °C.

The determination of specific properties on the plastic clay body and fired clay body were performed according to the standards listed in Table 4.

Table 4. The test method for measuring of selected properties of plastic clay body and brick clay body

Property	Standard
Working moisture content, wt.%	STN 72 1074
Linear drying shrinkage, wt.%	STN 72 1565-5
Linear change total, wt.%	STN 72 1565-5
Weight loss by firing, wt.%	STN 72 1565-5
Water absorption by boiling, wt.%	STN 72 1565-6
Apparent porosity, wt.%	STN 72 1565-6
Bulk density, kg/m^3	STN 72 1565-6

An ISOMET Model 104 (Applied Precision Ltd., Bratislava) was used to determine the thermal conductivity coefficient. Compression strength was determined on the samples dimensions of 20 x 20 x 20 mm. Porosity measurements (pore volume and median pore radius) were made using a high pressure mercury porosimeter (Thermo Finnigan Pascal 240, firm Thermo Scientific).

3. RESULTS AND DISCUSSION

The results obtained for the seven mixtures are shown in Table 5. Among the aforementioned properties, the most important are thermal conductivity and compressive strength, whose values are closely associated with the apparent porosity of the fired clay body (see Fig. 5).

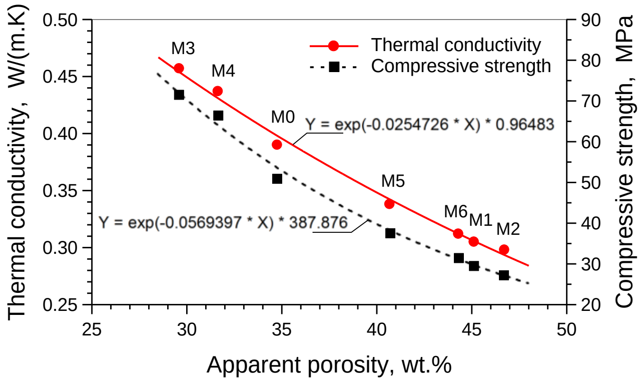

Fig. 5. The relationship between the apparent porosity and thermal conductivity / compressive strength

This relationship is expressed by a power function and it confirms the results obtained by [23]. In our case, the apparent porosity of the fired clay body depended on two important factors: the working moisture and the dose of combustible pore-forming agent. By applying Vuppor additive, we achieved a significant increase in the working moisture while maintaining a constant plasticity of the brick clay (mixtures of M1, M2, M5 and M6). Vuppor additive usage allowed for a simultaneous reduction of the linear drying shrinkage (M1, M2, M5 and M6), which led to a synergistic effect that positively affected pore structure growth and decreased the thermal conductivity when compared to mixtures M0, M3, and M4, see Table 5. However, an important consideration here is that the combustible agent also affected the thermal conductivity coefficient (M0, M1, M2, M4, M5 and M6). The results obtained by mercury porosimetry on the fired samples are shown in, Fig. 6 and Fig. 7. These results support the fact that we can easily achieve similar porosity levels when replacing greater part of combustible pore-forming agents with a small dose of Vuppor additive (M0 and M5). Our research was focused on assessing how pore size affects the thermal conductivity of the fired clay body. The results showing the effects obtained for pore size ranging from 0.001 to 0.1 µm are shown in Fig. 8, and those for pore size between 0.1 and 5 µm are shown in Fig. 9.

The results did not indicate a correlation between pore size ranging from 0.001 to 0.1 µm and the thermal conductivity (Fig. 8).

Table 5. Properties of the test samples before or after the firing process

Property	Test mixture						
	M0	M1	M2	M3	M4	M5	M6
Working moisture, wt.%	26.33	31.65	33.09	25.93	25.38	31.55	33.19
Linear drying shrinkage, wt.%	– 5.25	– 4.67	– 4.58	– 5.66	– 5.55	– 5.17	– 4.99
Linear change total, wt.%	– 4.88	– 4.52	– 4.71	– 5.39	– 5.3	– 5.09	– 5.08
Weight loss by firing, wt.%	12.96	13.09	13.11	10.83	11.42	11.67	11.86
Water absorption, wt.%	21.29	31.18	32.6	18.58	18.11	26.54	29.87
Apparent porosity, wt.%	33.76	45.12	46.72	31.64	30.6	40.68	44.3
Bulk density, kg/m^3	1585	1448	1433	1703	1689	1533	1483
Thermal conductivity, W/(m.K)	0.384	0.305	0.298	0.457	0.438	0.338	0.312
Compressive strength, MPa	51.2	30.0	27.1	81.5	69.0	37.7	31.5

Table 6. Increasing cost on the combustible pore-forming agents and additive Vuppor for the test mixtures per 1 t of the brick product

Testing mixture	**M0**	M1	M2	M3	M4	**M5**	M6
Price, €/t	**17.60**	26.10	30.40	-	5.90	**14.00**	18.30

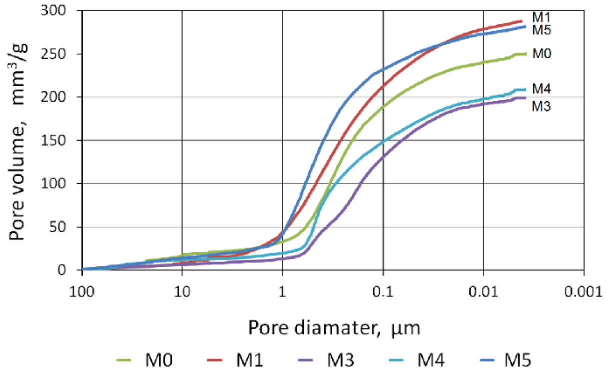

Fig. 6. Pore diameter distribution curves on the fired test samples

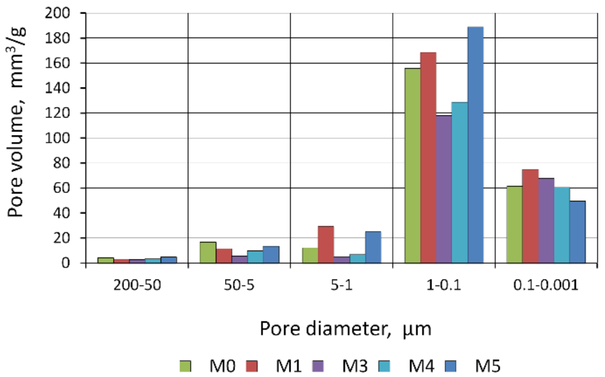

Fig. 7. Histograms of the distribution of the pore volume

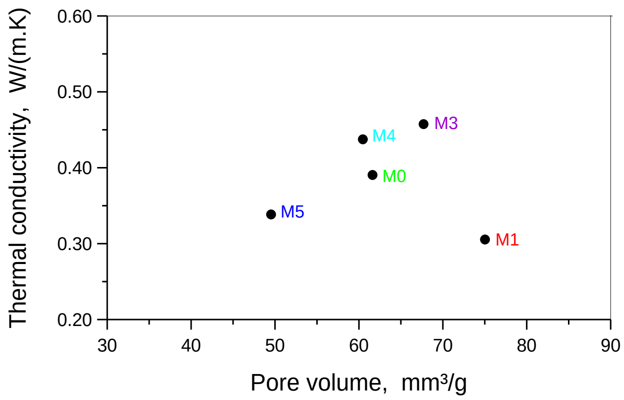

Fig. 8. Relationship between pore volume and thermal conductivity in pore size of 0.001 to 0.1 µm

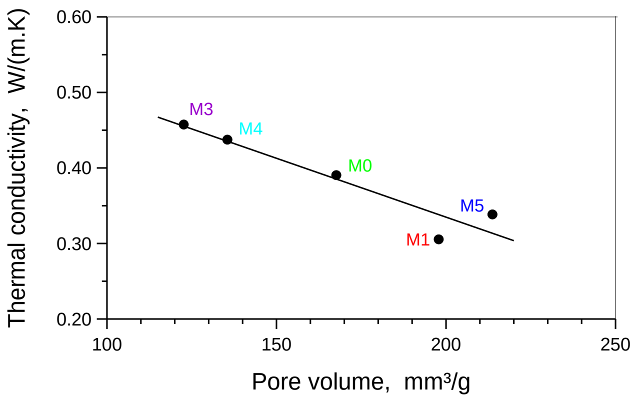

Fig. 9. Relationship between pore volume and thermal conductivity in pore size of 0.1 to 5 µm

In contrast, a linear regression existed between pore size ranging from 0.1 to 5 µm and thermal conductivity (Fig. 9). Based on this, it was confirmed that thermal conductivity is affected more by pore sizes above 0.1 µm than by pore sizes below 0.1 µm. Moreover, these results are consistent with the results of the researchers mentioned earlier [16 – 18].
On the other hand, price can be an important factor when selecting the most appropriate mixture for brick production (as shown in Table 6). Price of mixture per ton of brick was calculated based on the next input prices: sawdust 13.5 €/prms, coal sludge 75 €/t, Vuppor 0.8 €/kg. Based on the results listed in Table 6, by using mixture M5 (5 vol.% sawdust + 0.5 wt.% Vuppor additive) thermal conductivity can be decreased from 0.384 to 0.338 W/(m.K) and production costs can be decreased by 20 % when compared with the mixture currently used (M0).

Based on the above values of CO_2 emissions from burning 1 kg pore-forming agents we calculated that in this case CO_2 emissions can be reduced by 60 %. Further reduction of CO_2 emissions can be achieved through shortening of the burning process and through reduction of the firing temperature [20, 21].
On the other hand, conventional combustible pore-forming agents can be used, for example in the furniture industry, the biogas production, the energy sector - fluidized bed combustion, the waste composting methods or the bioethanol production that could be a more environmentally friendly solution (for reducing CO_2 emissions) than the actual ones.

4. CONCLUSIONS

1. Nowadays, brick industry is characterized by the use of high amounts of combustible pore-forming agents like sawdust and cellulose and coal sludge, which are added to increase the porosity of the fired clay body. This trend was caused by customers who prefer products with high thermal insulation properties. It is also known, that conventional pore-forming agents are gradually more expensive and they are also causing several problems in the production of bricks, but above all they increase the production of CO_2 emissions. Our research results show that a pore percentage increase in the fired clay body can be achieved not only by applying combustible pore-forming agents but also by the application of Vuppor additive.
2. Vuppor additive, for example, has the same effect on pore structure formation on the fired clay body than conventional pore-forming agents while reducing CO_2 emissions.
3. The presence of Vuppor additive in the fired clay body increases the proportion of pores, especially with a size range between 0.1 and 5 μm, having a positive impact on the reduction of its thermal conductivity in the application of brick raw material with a high proportion of carbonates.
4. By using a dose of 0.5 wt.% Vuppor additive, the production costs can be reduced by 20 % and the thermal conductivity by 12 % and CO_2 emissions can be reduced by 60 %.

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