



Development of extruded and fired bricks with steel industry byproduct towards circular economy



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ARTICLE INFO

Article history:

Received 2 May 2016

Received in revised form

10 July 2016

Accepted 16 August 2016

Available online 17 August 2016

Keywords:

Building bricks

Steel industry byproduct

Extrusion

Firing

Characterization

ABSTRACT

In the present research, the development of building bricks is examined, using steel industry electric arc furnace dust (EAFD) as admixture into standard clayey raw materials typically used by ceramic industries, and employing a pilot-plant simulation of industrial processes for red brick manufacturing. The recycling of solid residues, which are derived in massive quantities from steel production plants, as alternative raw materials towards circular economy, is of increasing importance. In particular, steel dust recovered from EAF gas treatment, contains several oxides and, thus, can be considered as secondary material for substituting clays in traditional brick manufacturing. Possible economic benefits for the energy intensive industrial ceramic sector from energy savings upon firing along with a high potential for environmentally safe management of steel dust should be emphasized. For that purpose, various clay/EAFD mixtures were prepared and mixed with water to form a plastic mass for brick specimen shaping by extrusion. The green specimens were dried, and then fired at different peak temperatures (850, 950 and 1050 °C) in a controlled laboratory chamber furnace for sintering and consolidation. The effect of the by-product content (%) and of the firing temperature on brick shrinkage, bulk density, water absorption capability, mechanical strength and thermal conductivity was investigated. According to the results, the development of extruded and fired bricks with up to 15 wt% recycled steel industry byproduct is feasible without significant variations in their technological properties.

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1. Introduction

The aim of "closing the loop" of product lifecycles through greater recycling, by safely turning waste byproducts from an industry into useful secondary resources for another industrial sector is strongly encouraged by current European Union policies, towards industrial symbiosis, ample coordination and circular economy.

Huge quantities of clays are annually needed for the production of considerable amounts of fired ceramic bricks worldwide, and therefore much research focuses on the utilization of alternative raw materials from various origins into clay mixtures, at different combinations and proportions, for the fabrication of conventional sintered bricks [1–3].

On the other side, management and valorization of massive quantities of solid residues recovered in steelmaking plants worldwide, such as electric arc furnace dust, electric arc furnace slag and ladle furnace slag, represent a significant issue. In steel industry, the production of 1 ton of steel results in generation of 2–4 ton of various types of waste by-products [4], while 1 ton of

stainless steel waste is produced per 3 tons of stainless steel-making [5]. Taking into account the quantities of steel by-products, their proper disposal and handling remains both dangerous and expensive task, and therefore, their safe utilization can be environmentally and financially beneficial. Blast furnace slag and steel slag are already competitive raw materials in the mineral industry, and blast furnace slag use in the cement industry currently increases, resulting in production cost reduction. Also, electric arc furnace slag is widely used in the road and pavement construction and is recently studied for the development of vitreous ceramic tiles [4,6,7].

Especially, the recycling of electric arc furnace dust (EAFD) is very important, because it is one of the major steel by-products, included in the European Waste Catalogue [8], and produced worldwide in large quantities [9–12]. EAFD is generated from the volatilization of heavy metals when steel scrap is melted in the electric arc furnace. Volatilized metals are oxidized and subsequently solidified and detained in the form of fine powder in specially designed filters, which are placed in the EAF gas stream cleaning system [13–16]. The use of EAF technology in the steel-making industry has been increasing considerably over the last decades, resulting in the production of significant quantities of solid residues. The world generated steel dust per year has been

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estimated to be around 3.7 million tons [17].

EAFD mainly contains zinc and other metals, and it is recycled in USA for recovering zinc and lead from the industrial waste stream, employing the Waelz Kiln technology. Main product is Waelz oxide, which is transformed into zinc oxide, zinc sulfate or zinc metal by zinc smelters. Waelz iron product, an iron concentrate, is also produced [18]. Moreover, steelmaking dust is examined as an additive in asphalt cement mixtures for road construction [19]. Environmentally friendly activities aiming at the recovery of massive quantities of steel dust generated, through various forms of recycling and utilization, are in accordance with EU regulations [20].

Since EAFD contains several useful metal oxides, it can be considered as secondary raw material for substituting traditional clayey minerals in ceramic manufacturing. Given its iron content, it can be used to improve the ceramic product color. From the environmental point of view, the recycling of steel-industry EAFD as secondary raw materials in brick production would contribute to conservation of natural resources. Furthermore, the low cost of this largely available solid waste and even possible energy savings during clay/waste mixtures firing in brick manufacturing should also be considered, particularly taking into account recent targets of the EU's energy policy [21].

Recent studies reported in the literature reveal an increasing interest on the utilization of steelmaking dust in the fabrication of construction materials including ceramics, glass-ceramics, vitreous products and geopolymers [22]. Specifically, the possibility for using EAFD along with waste glass into compacted glass-ceramics is promoted by alkalis present in dust, which can act as fluxing constituents [23]. On the other hand, mixing 10 wt% steelmaking dust with fly ash for preparing geopolymers provides the highest value of compressive strength, while amounts of more than 10 wt% dust lead to rapid decrease of the compressive strength [24]. Particularly for ceramics, it was determined that adding steelmaking dust up to 5 wt% into the brick mass imparts slight increase of compression strength (at sintering temperatures of 950 °C and 1150 °C) [20]. Moreover, EAFD was indicated to act as a non-plastic material that could be incorporated into two types of clays (one from Argentina and another from Brazil) to adjust their workability for shaping compacted ceramics. In this case, it was found that recycling up to 20 wt% of steel dust into red ceramics fabricated by compression is technologically feasible [25]. Also, blend of clay with up to 20 wt% EAFD (fired in the range of 800–1100 °C) yielded compacted ceramics appropriate for use in structural applications [26]. Besides, EAF dust-based bricks containing 15 wt% bentonite as admixture and held for 30 min at 1000 °C that can be useful as thermal insulators in non-ferrous reactors and allied devices were also prepared [27]. Furthermore, in a synthetic mixture of clayey powders containing the minerals biotite, muscovite, montmorillonite as well as quartz and feldspars, up to 20 wt% steel dust was added to manufacture extruded clay-based ceramic products (fired at 850–1050 °C) [28].

In the present research, plastic extrusion procedure was employed for shaping standard clayey raw materials typically used by Greek ceramic industries for the formation of traditional clay bricks, loaded with as much steel industry byproduct (electric arc furnace dust, a non-plastic secondary resource) as possible into brick specimens, using a pilot-plant simulation of industrial processes for red brick manufacturing. The effect of the admixture percentage as well as of the firing temperature on technological properties of the extruded and fired bricks obtained after sintering is examined.

2. Materials and methods

2.1. Raw materials

Three clay samples from different deposits in Greece, representative of the main types of clayey raw materials utilized by the ceramic industry (A, B and C), were selected and characterized (XRF-analysis). Their chemical composition is shown in Table 1. It can be seen that the CaO content of the clay samples used ranges from 3.51 to 11.82 wt%. No sulphur was detected. The main mineralogical phases identified were albite ($\text{NaAlSi}_3\text{O}_8$), enstatite ($(\text{Mg, Fe})\text{SiO}_3$) and illite.

In Table 2, the chemical composition of the steelmaking dust (EAFD) sample (red/brownish fine powder), which was used as a secondary raw material in the current, is presented. From Table 2, it is evident that iron and zinc are the main EAFD constituents. This is in accordance with several other studies on the characterization and management of steel-industry dust. Zinc originates mainly from scrap recycled in electric arc furnace processes. The dust generated and collected is usually classified as EAFD with a high (> 15%) zinc content or with a low (< 15%) zinc content [22]. Each particular steelmaking dust is certainly site-specific. The zinc typically exists in the dust as zinc oxide (ZnO) and as a mixed zinc-manganese ferrite spinel or ZMFO ($(\text{Zn}_x\text{Mn}_y\text{Fe}_{1-x-y})\text{Fe}_2\text{O}_4$) [29].

2.2. Specimen preparation and testing

Specimens having dimension $80 \times 43.5 \times 18$ mm were prepared employing a pilot-plant simulation of the industrial brick manufacturing processes: the clay samples were pulverized and mixed in proportions appropriate for standard brick fabrication. Various clay/EAFD mixtures with 0–15 wt% dust content were prepared and mixed with water to form a plastic mass for extrusion. After gradually drying a) in air for 24 h and subsequently b) in an oven at 105 °C for 48 h, the specimens were fired in a chamber furnace for sintering and final consolidation. The first heating step of 500 °C was reached after controlled heating at 1.7 °C/min for 5 h, followed by further heating at 4.5 °C/min up to a peak temperature. The specimens remained at the maximum temperature only for 15 min in order to attain energy savings, and then they were cooled to room temperature in the furnace.

After optimizing the % EAFD content in the mixture by firing at 1050 °C (at higher sintering temperatures also tested, the brick bodies appeared to be over-fired), similar heating procedure was followed by lowering the maximum firing temperature to 950 °C and also down to 850 °C, to assess the possibility for achieving energy savings.

Phase identification was conducted via X-ray diffraction (XRF)

Table 1
Chemical composition (wt%) of the clayey raw materials.

Composition	Type of clay		
	A	B	C
SiO ₂	46.88	52.64	47.67
Fe ₂ O ₃	5.61	5.22	3.82
Al ₂ O ₃	17.98	19.05	9.55
TiO ₂	0.72	0.68	0.43
CaO	7.53	3.51	11.82
MgO	8.15	5.91	8.39
SO ₃	0.01	0.00	0.00
P ₂ O ₅	0.14	0.18	0.98
Na ₂ O	1.75	2.45	2.03
K ₂ O	3.15	3.65	1.36
LOI*	7.5	6.4	12.8

* LOI : Loss on Ignition

Table 2
Main components (wt%) of the EAFD powder used.

Component	Composition (wt%)
FeO	37.7
ZnO	22.0
PbO	12.0
CaO	7.7
Na ₂ O	5.1
SiO ₂	3.7
MnO	3.2
MgO	2.2
K ₂ O	2.1
SO ₃	1.4
Cl	1.3
Al ₂ O ₃	0.7

measurements. Shrinkage upon sintering as well as bulk density, open porosity, water absorption capability (%), mechanical strength and thermal conductivity of sintered specimens were determined and studied in relation to the admixture percentage and also to the firing temperature.

In order to determine the water absorption capacity and open porosity, the sintered specimens were weighed before and after immersion in water for 24 h (Eqs. (1) and (2) respectively):

$$WA(\%) = 100(W_{\text{wet}} - W_{\text{dry}})/W_{\text{dry}} \quad (1)$$

$$OP(\%) = 100(W_{\text{wet}} - W_{\text{dry}})/(\rho V_s) \quad (2)$$

where:

W_{wet} = specimen weight when saturated with water (g).

W_{dry} = specimen weight when dry (g).

ρ = density of water (1 g/cm³).

V_s = specimen volume (cm³).

Mechanical behavior was assessed by three-point bend testing (flexural strength). Tests were performed on 20 specimens of each composition and firing temperature, and the average values were reported in the results. Then, the modulus of rupture (M.O.R.) was calculated from the following relationship (Eq. (3)):

$$M.O.R.(\text{MPa}) = (3PL)/(BW^2) \quad (3)$$

where:

P = the fracture load (N).

L = half of the span between the supports of the bend ring (mm).

B = the width of the specimen (mm).

W = the height (thickness) of the specimen (mm).

The thermal conductivity coefficient (k) was measured at 25 °C using the guarded heat flow meter method (Anter Unitherm Model 2022) in accordance with the ASTM E1530: Standard Test Method for Evaluating the Resistance to Thermal Transmission of Materials by the Guarded Heat Flow Meter Technique.

3. Results and discussion

3.1. Brick specimen preparation

The increasing incorporation of EAFD into the clay raw materials reduced the plasticity of the mixture. Nevertheless, the workability of the plastic mass obtained up to 15 wt% EAFD addition did not cause significant problems to the extrusion of integral green (unfired) specimens possessing the strength required to ensure safe handling in the subsequent fabrication stages. Therefore, the extrusion behavior of the green bodies was acceptable, and no additive was demanded to facilitate the plastic extrusion of clay mixtures loaded with up to 15 wt% steel dust. Generally, the embodiment of industrial solid by-products into clay bricks produced by extrusion has rarely exceeded a 20–30 wt%, due to a certain incompatibility of the industrial residues with the clay mixtures during the production steps, especially regarding a reduced plasticity at higher admixture percentages.

The behavior of green specimens during heating was quite satisfactory. Coloring of fired brick specimens turns gradually from lighter to darker brown when the EAFD percentage in the raw material mixture is increased, due to its noticeable % iron content.

Typical XRD spectra of as-received EAFD as well as of 10 wt% EAFD-loaded clay bricks sintered at 850 °C and 1050 °C are provided in Fig. 1. Iron-rich phases are clearly present. Phase evolution with firing temperature involves oxidation of Fe₃O₄ (contained in EAFD) into Fe₂O₃, which is favored by the sintering temperature increase up to 1050 °C, for oxidizing conditions [30]. In fact, vigorous calcining of magnetite (Fe₃O₄) in air gives hematite (Fe₂O₃) (Eq. (4)):

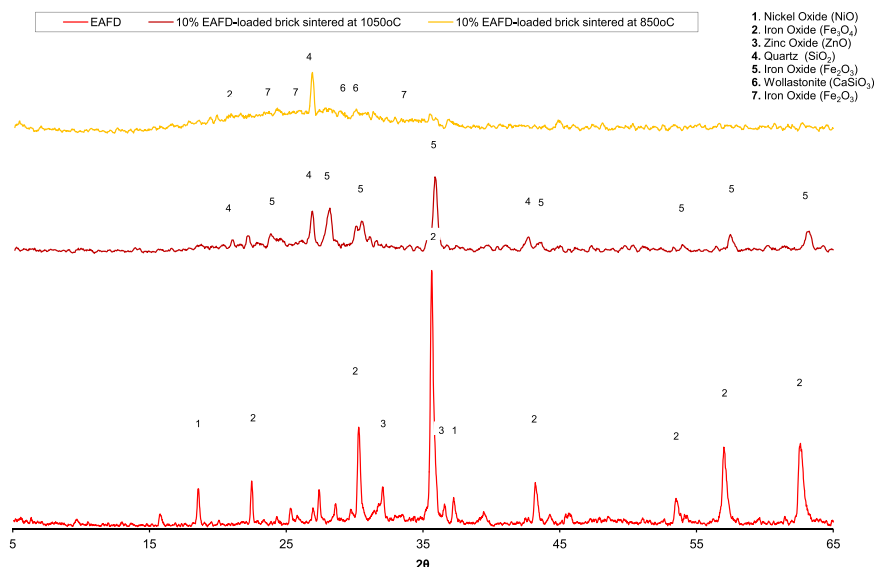


Fig. 1. Typical XRD spectra of as-received EAFD and also of 10 wt% EAFD-loaded clay bricks sintered at 850 °C and 1050 °C.

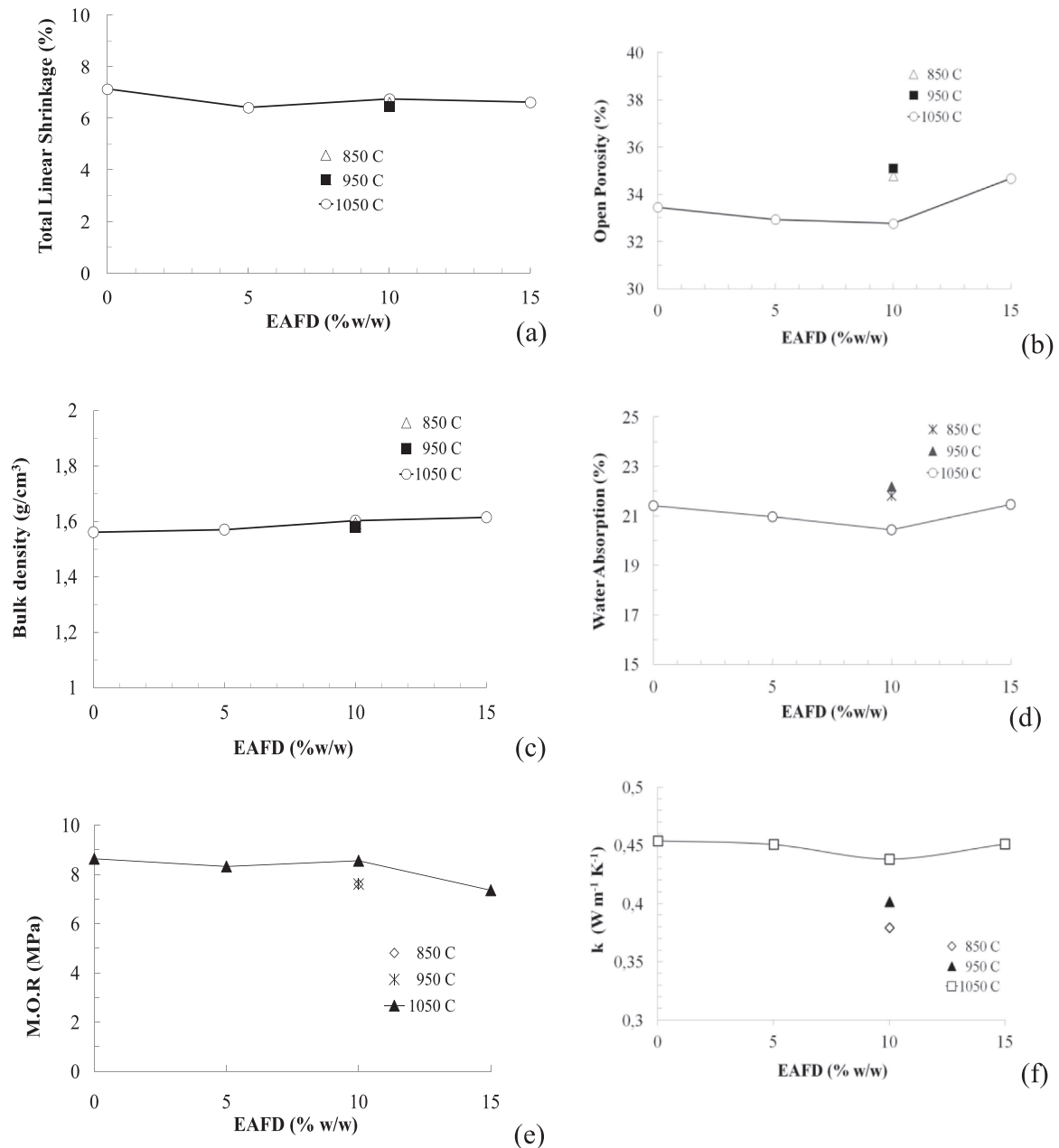


Fig. 2. Effect of the EAFD addition and also of the firing temperature on shrinkage (a), open porosity (b), bulk density (c), water absorption capacity (d), modulus of rupture (M.O.R.) upon 3-point bend testing (e) and coefficient of thermal conductivity - k (f) of sintered specimens.



Besides, a more pronounced crystallinity is obtained when raising the sintering temperature from 850 to 1050 °C.

The physico-mechanical properties measurements results obtained in this study for EAFD-loaded brick specimens are compared to reference values of prototype clay brick specimens (0% EAFD) produced under similar experimental conditions with similar clayey raw materials, as follows:

3.2. Effect on physical properties

The influence of the EAFD addition into clay-based specimens fired at 1050 °C, as well as of the firing temperature for 10 wt% EAFD-loaded specimens, on the total linear shrinkage of green specimens upon heating is depicted in Fig. 2a, and also on the open porosity and the bulk density of sintered specimens in Fig. 2b

and c respectively. It can be seen from Fig. 2a that shrinkage appeared relatively restricted in all cases, and remained within tolerable limits for standard brick manufacture. Fig. 2b shows that open porosity and bulk density do not vary significantly with the % EAFD content. The lower open porosity is determined when 10 wt% EAFD is added into the clay mixture sintered at 1050 °C, which can be attributed to the most intense fluxing action of steel dust at these conditions.

Water absorption capability can restrict the utilization of ceramics as building materials. Ceramic products used as flooring should have a water absorptivity < 1%, as roofing tiles < 20%, and as hollow bricks about 25% [26]. The results for the water absorption of all sintered bricks produced in the present study lie in the range of approx. 20–23% (see Fig. 2d). The trend in water absorption results appears generally similar to that for the open porosity (Fig. 2b). Specifically, the water absorption of bricks (sintered at 1050 °C) slightly decreases, as the EAFD amount in the

mixture is increased up to 10%, following the decrease in porosity already mentioned. Further waste addition up to 15 wt% leads to a slight increase in water absorption. Regarding the firing temperature effect (on 10% EAFD specimens), it can be seen that water absorption capacity remains almost constant, when the sintering temperature is increased from 850 to 950 °C, but it clearly decreases when firing the specimens at 1050 °C, as a result of better consolidation, with the beneficial fluxing action of dust, at this firing temperature.

The effect of the EAFD addition into the clay mixture for specimens fired at 1050 °C, as well as of the firing temperature for specimens containing 10 wt% EAFD, on the brick bulk density is presented in Fig. 2c. According to the results, bulk density of sintered specimens is only slightly affected by the % EAFD content and the firing temperature. These findings are also in accordance with the experimental results for the weight loss upon sintering, also determined, which does not vary substantially and lies approximately in the range of 9.5–10.5%.

3.3. Effect on mechanical strength

The effect of the EAFD addition into the clay mixture for specimens fired at 1050 °C, as well as of the firing temperature for specimens containing 10 wt% EAFD, on flexural strength, expressed in terms of modulus of rupture (M.O.R.) calculated upon three-point bending of the bricks (Eq. (3)), is shown in Fig. 2e.

The experimental data indicate that addition of up to 10 wt% electric arc furnace dust into the clay mixture, has no opposite effect on the M.O.R. of sintered bricks. So far, slight improvements in compression strength of compacted have been reported by other researchers only for 5 wt% steel dust addition in the ceramic bodies. It should be noted, however, that further waste addition (15 wt%) into the clay mixture leads to a noticeable decrease of approx. 15% in M.O.R. due to the already stated more pronounced porosity at this EAFD percentage. In this case, it seems that the amount of admixture that can act as a fluxing agent in the ceramic mass is overpassed.

With regard to firing temperature effect, no difference in M.O.R. is observed for a temperature increase from 850 to 950 °C, but the M.O.R. increases by approx. 12% when firing the specimens at 1050 °C. It should be noticed, here, that strength of sintered materials is strongly dependent on their porosity. Flexural strength in particular, is reported to take on an exponential reduction with increasing porosity, according to Rice formula [31] (Eq. (5)):

$$\sigma = \sigma_0 \exp(-bp) \quad (5)$$

where

σ : the strength of a microstructure with a porosity (p)

σ_0 : the strength of the non-porous structure

b : an empirical constant

Hence, the improvement in M.O.R. for bricks sintered at 1050 °C should be attributed to the aforementioned decrease in open porosity, associated with an enhanced consolidation, at this sintering temperature.

Temperatures above 950 °C lead to higher M.O.R. values, but can also involve difficulties in holding mortar to the bricks, as it was reported by other researchers [32].

3.4. Thermal conductivity

Use of ceramics as thermal insulators represents one of the main applications for this category of materials. The usefulness of a ceramic for these applications is largely fixed by the rate of heat transfer through it under a particular temperature (T) gradient. The basic equation (Eq. (6)) to define the thermal conductivity

coefficient (k) is [33]:

$$dQ/d\theta = - (kAdT)/dx \quad (6)$$

where:

dQ = the amount of heat flowing normal to the area A in time $d\theta$

$-dT/dx$ = the temperature gradient

k = the thermal conductivity coefficient, the proportionality factor, a material constant.

Fig. 2f shows how k of sintered bricks is affected by a) the % EAFD into the clay mixture (at 1050 °C) and b) the firing temperature (for 10% EAFD-loaded specimens). It is apparent that k does not vary considerably with the EAFD addition up to 15% (1050 °C) and remains relatively constant (around $0.45 \text{ W m}^{-1} \text{ K}^{-1}$). On the other hand, k increases when the sintering temperature for 10 wt% EAFD specimens is increased from 850 °C to 950 °C, and especially up to 1050 °C. This variation in ceramic thermal conductivity should mainly be attributed to the corresponding decrease in open porosity [34].

4. Conclusions

- Clay-based bricks are successfully developed by plastic extrusion and firing using recycled steel industry byproduct as admixture, and employing pilot-plant simulation procedures of industrial brick manufacturing.
- Effective extrusion of brick specimens incorporating up to 15 wt% EAFD into the clay mixture is feasible, without significant variations in both their mechanical performance (flexural strength, in terms of M.O.R.) and thermal conductivity, after sintering. Further % admixture use would endanger the extruded product quality. EAFD practically does not act as pore-forming agent in the bricks produced, as only slight increase in the open porosity occurs, even with 15 wt% dust addition into the clay raw material mixture.
- When the firing temperature is increased up to 1050 °C, M.O.R. and thermal conductivity (for 10 wt% EAFD-loaded bricks) slightly increase as a result of better densification and higher crystallinity.

Acknowledgment

This research has been co-financed by the European Union (European Social Fund - ESF) and Greek national funds through the Operational Program "Education and Lifelong Learning" of the National Strategic Reference Framework (NSRF) – Research Funding Program: ARCHIMEDES III. Investing in knowledge society through the European Social Fund (Scientific Coordinator: Assoc. Professor Vayos G. Karayannis).

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