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Using mordenite-rich tuff as a natural clay replacement in fired clay brick production

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Abstract

This work examines the impact of mordenite-rich tuff (MT) on the technological properties of fired clay brick. These specimens were obtained by substituting clay with MT in various proportions (0, 5, 10, 15, 20, 50, and 100 wt%). They were pressed at 10 MPa, air-dried in the laboratory, and fired at 900 °C using two different clays: clay of Remila (CR) and clay of Adjiba (CA), Algeria. The mechanical, thermal, and physical properties of fired bricks were determined and compared to those of control bricks (0 wt%). According to the finding, the partial and total substitution of clay by MT reduced the sample's thermal conductivity and bulk density. Thus, the MT increased the apparent porosity and the water absorption of clay brick while maintaining the compressive strength within the limits of the Algerian standards. This research revealed the possibility of producing bricks containing up to 100% by weight of MT with the required technical and environmental properties.

Keywords: mordenite, tuff, clay, pores, compressive strength, fired brick.

INTRODUCTION

In recent years, brick production has grown significantly around the world, with an annual global production of around 1.5 trillion clay bricks produced each year [1]. Bricks are increasingly preferred as a building material because of their economical production and quick laying [2]. This requires the extraction of a significant amount of clay, used as a raw material in the production of clay brick, about 3.13 billion m^3 of clay soil per year [3]. This leads to a gradual depletion of the natural reserves of this product. In order to preserve clay resources and environmental balance, it is important to find alternative materials to replace clay in bricks without affecting the brick's technical properties [4]. The use and suitability of a specific material depend mainly upon its availability, and technical, environmental, and economical considerations. Many researchers have therefore focused on the development of brick material technology using waste by-products to reduce the consumption of natural clay from the soil. Waste glass [4], sewage sludge [5], fly ash [6] and quarry tailings [7] have all been used to replace clay in brick production.

Natural zeolites are hydrated, crystalline microporous aluminosilicates, containing alkaline earth metals and alkali, and are mainly formed by the weathering of volcanic ash, which is a fine amorphous rock rich in SiO_2 and Al_2O_3. They are mineral rocks with a chemical composition similar to that of clays. The nature of geological deposits and their mineralogical composition have a direct impact on the physicochemical properties of zeolites, which, by their variations, enable the use of the zeolite in various technological processes [8]. Scientific knowledge and different applications of natural zeolites are constantly developing owing to the introduction of new resources or uses. Nagrockiene and Girskas [9] studied the characteristics of concrete modified by the addition of natural zeolite. Ulloa et al. [10] recently used natural zeolite in geopolymer mortar formulations. Mertens et al. [11] studied the pozzolanic reactions of natural zeolites with portlandite and the factors influencing their reactivity. Perraki et al. [12] studied the effect of zeolite on the properties and hydration of mixed cements. Meziani et al. [13] determined the effects of replacing cement with mordenite-rich tuff (MT) on the transport properties and life of cementitious materials. Vakalova and Revva [14] recently used natural zeolite in the production of brick. They found that adding zeolite rock (10 to 30 wt%) in the production of ceramic brick functions as a humidity extractor. Ibrahim et al. [15] studied the properties of bricks manufactured from sawdust and zeolite-poor rock. They concluded that a zeolite-poor rock with 8% sawdust is the best combination for making high-quality composite bricks. Although zeolitic materials have been the subject of several research studies on their usage as construction materials, relatively few studies have been done to evaluate them as raw materials for brick production. To the best of our knowledge, no research has been done to examine the use of mordenite-rich tuff as the main raw material for making fired bricks, and the physical properties like bulk density, water absorption, porosity, compressive strength, and thermal conductivity for mordenite-rich tuff in the production and characterization of fired brick have not been explored.

The main objective of this study is to investigate the possible use of zeolitic material as an alternative and environmentally friendly building material. In order to

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preserve natural clay by substituting it with MT, this study examined the effects of MT with partial and full clay substitution rates on the characteristics of clay brick. It also offers the brick industry a new, light material with reinforced insulating properties and lower CO_2 emissions. Investigations were made on the impacts on the technical characteristics of fired bricks, such as compressive strength, water absorption, apparent porosity, bulk density, and thermal conductivity.

MATERIALS AND METHODS

Two types of clay were utilized in the production of bricks: the first, grey in color, came from a quarry in Remila (CR), and the second, red in color, came from a quarry in Adjiba (CA), both in Algeria. The mordenite-rich tuff (MT) came from natural deposits in the Tinebdar area (Bejaia) in Algeria. The clay was first sun-dried for 72 h, then dried in an oven at 105 °C for 48 h before sieving into a 2 mm sieve. The MT, white in color, was sun dried before being placed in a 105 °C oven for 48 h before crushing and sieving into a 2 mm sieve. The identification of the mineralogical phases in the raw materials (CA, CR, and MT) was determined by the X-ray diffraction (XRD) technique (D5000, Siemens). The chemical characteristics of the different clays and tuff were determined by X-ray fluorescence spectroscopy (XRF, SRS 300, Siemens). The particle size distribution of the materials was determined by using a laser scattering particle size distribution analyzer (Horiba). The real density was determined using a helium pycnometer (AccuPyc II 1340, Micromeritics).

The brick specimens were obtained by mixing X= 0, 5, 10, 15, 20, 50, and 100 wt% of MT with 100-X of clay (CR or CA). An optimum amount of water of 8-12% was added to the CA-MT mixtures, and an amount of 9-12% was added to the CR-MT mixtures. In a 5 L mixer, the MT was added to the clay and dry mixed for 3 min to obtain a homogeneous material, and then water was gradually added to the dry mixture before it was mixed again for 2 min. Following homogenization of the fresh material, a uniaxial hydraulic press (KCK-30, Mega) was used to press the mixture into the molds (for mixed formulations, a constant 10 MPa pressure was used). The samples were first kept in a storage room at a controlled temperature of 20 °C for 72 h then dried in an oven at 105 °C until a constant mass was obtained. Finally, dried samples were fired in a kiln with an initial slow heating rate of 2 °C/min up to 600 °C, then 4 °C/min up to 900 °C and dwelled for 2 h at the maximum temperatures. The manufactured clay bricks were then subjected to a series of tests according to the standards. The molds' dimensions were 40x40x160 mm^3 for measuring physical properties, 40x40x40 mm^3 for measuring mechanical characteristics, and 20x80x50 mm^3 for measuring thermal properties (5 samples in each series).

The microstructures of the samples were observed with a scanning electron microscope (SEM, SM 840, Jeol) equipped with an energy dispersive spectroscope (EDX) for microchemical analysis (20 kV). The samples were placed on aluminum support and carbon coated with an ion sputter (JFC 1100, Jeol). Bulk density and apparent porosity of fired clay bricks were determined following ASTM C 373-88 standard [16]. Industry-standard method UNE-EN 772-21 was used to calculate water absorption [17]. The compressive strength of the fired brick was measured with a testing machine (65-L11D2, Controls) applying the load centered on the surface of the brick at 20 MPa/s until rupture. According to NFE 993-15 standard [18], the thermal property of fired brick was determined by using the hot wire method using the commercial CT-meter device (according to the Scientific and Technical Center for Building) [19].

RESULTS AND DISCUSSION

Physicochemical characterization of raw materials

The particle size distribution curves of CR, CA, and MT used in this study are shown in Fig. 1. CR and CA clays were finer than MT, which was justified by the low SiO_2 content of the two clays compared to MT (Table I). The CR and CA clays contained, respectively, 84% and 87% particles below (20 μm) and about 16% and 13% fractions above 20 μm. The fraction >20 *μ*m consisted of quartz and muscovite, while the particle size fraction <20 μm was composed of a mixture of quartz, muscovite, kaolinite, palygorskite, and gypsite. Only about 48% of the MT particles passed the 20 μm sieving. This showed that the MT powder had the largest fraction of grains with a diameter greater than 20 μm, which was due to the large proportion of SiO_2. This is important because it has been found that the gradation of the raw material influences the porosity of the brick after firing [20]. Moreover, the density of MT, 2290 kg/m^3, was lower than the density of CR and CA clay, which were 2645 and 2783 kg/m^3, respectively, indicating that lighter bricks can be obtained by replacing clay with MT. During the heating process, the oxides K_2O, Na_2O, Fe_2O_3, CaO, and MgO can improve vitrification and tend to result in a more

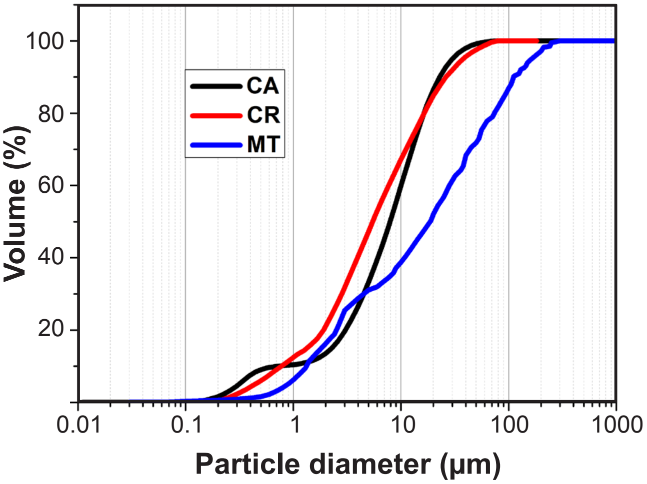

Figure 1: Particle size distribution of CA, CR, and MT.

Table I - Chemical composition (wt%) of CA, CR, and MT.

Raw material	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	K_2O	Na_2O	TiO_2	P_2O_5	LOI
MT	64.34	15.72	2.71	2.91	0.48	0.15	3.08	3.69	-	-	6.92
CR	43.30	13.35	5.01	15.34	1.89	0.71	2.03	1.06	0.63	0.19	16.50
CA	48.29	18.78	6.95	6.75	2.65	0.56	2.17	0.69	0.83	0.21	12.10

LOI: loss on ignition.

compact and dense brick structure, which is not ideal for enhancing the material's thermal behavior. As can be seen in Table I, MT contained a small amount of fluxing agents compared to either of the clays. This could make it possible to maintain the MT framework and improve the brick's thermal insulation.

XRD patterns of the raw materials are shown in Fig. 2. According to the XRD diffractogram of MT, mordenite was the main mineralogical phase present associated with heulandite since albite, muscovite, and sanidine are accessory minerals that affect the color of the tuff because they contain transition metals such as iron. The CA diffractogram revealed the presence of quartz, kaolinite, calcite, albite, muscovite, and dolomite. The CR diffractogram revealed the presence of calcite, muscovite, quartz, kaolinite, palygorskite, clinochlore, dolomite, gypsum, and albite. This result correlated well with that of the chemical composition (Table I) [21]. The chemical analysis of MT (Table I) showed that SiO_2 and Al_2O_3 had the highest major oxide contents, 64.34% and 15.72%, respectively. The sum of SiO_2 and Al_2O_3 constituted more than 80%, which was suitable for mechanical strength. Other oxides such as K_2O and CaO (5.99 wt% for each oxide) were linked with mordenite and heulandite. On the other hand, Fe_2O_3 (2.71 wt%) and MgO (0.48 wt%) were associated with muscovite, albite, and sanidine (Fig. 2). Both clays showed a similar composition, mainly consisting of silica (43.30% and 48.28% for CR and CA, respectively) and alumina (13.35% and 18.78% for CR and CA, respectively). A higher amount of CaO was noticed in CR. Loss on ignition value of raw materials was measured to be 6.92%, 16.5%, and 12.10% for MT, CR, and CA, respectively.

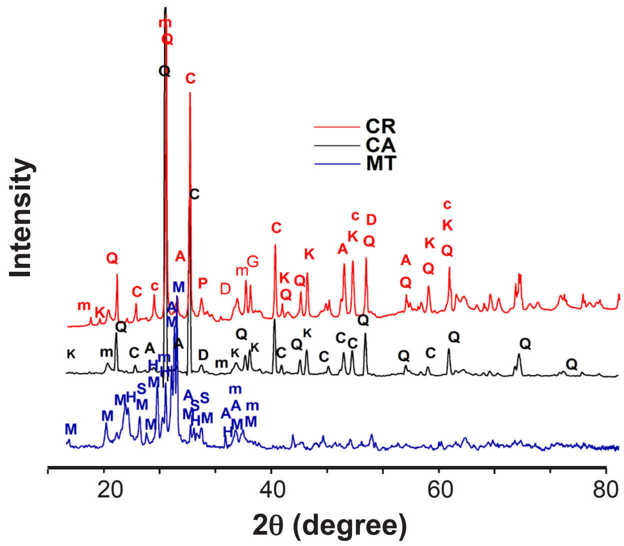

Figure 2: XRD patterns of the raw materials (CR, Ca, and MT). S: sanidine, H: heulandite, M: mordenite, A: albite, m: muscovite, Q: quartz, C: calcite, K: kaolinite, D: dolomite, P: palygorskite, C: clinochlore, G: gypsum.

Physical, mechanical, and thermal characteristics of the fired bricks

Apparent porosity: the porosity values of fired clay bricks are given in Fig. 3a. The increase in the substitution rate of clay by MT led to an increase in the apparent porosity of the bricks. Control bricks with CA and CR had an apparent porosity of 26.14% and 28.82%, respectively. The apparent porosity increased as the MT content increased to achieve a maximum value of 35.68% at a replacement level of 100%. This is explained by the MT porous structure (Fig. 4). MT is a molecular sieve because of its microporous nature. Nevertheless, since the fluxing agents had not yet melted to produce a sufficient glassy phase and because less liquid phase formed to fill the macro and micropores embedded in brick matrices, the bricks containing MT were not effectively vitrified at the firing temperature of 900 °C [22]. Because MT had a high silica concentration (i.e., more than 60%), the cohesiveness between the particles was destroyed, making the brick brittle [23]. This is explained by the fact that the addition of MT reduced the paste's cohesiveness by weakening its plastic characteristics and increasing the pore size within the brick [24]. This was confirmed by the bulk density and water absorption measurements given in Figs. 3b and 3c.

Water absorption: the ability of bricks to absorb water determines how long they will last. Less water infiltration indicates greater brick longevity and resilience to environmental deterioration. Water absorption is a key indicator of the degree of densification and its relationship with porosity values [25]. The absorption test results given in Fig. 3b show that raising the substitution rate of clay by MT increased the water absorption of the bricks. This can be attributed to the high absorption capacity of MT. Increasing the MT replacement increased the porosity of the brick, which increased water absorption. All bricks had water absorption of less than 22%. According to ASTM C 62-17 standard [26], all bricks could be used in moderate climate conditions or normal climate conditions except bricks with a substitution rate of less than 15% of CA and 10% of CR, which can even be used in severe weather conditions with water absorption of less than 17%. To prevent water incursion, the bricks' interior structure must be sufficiently dense [27].

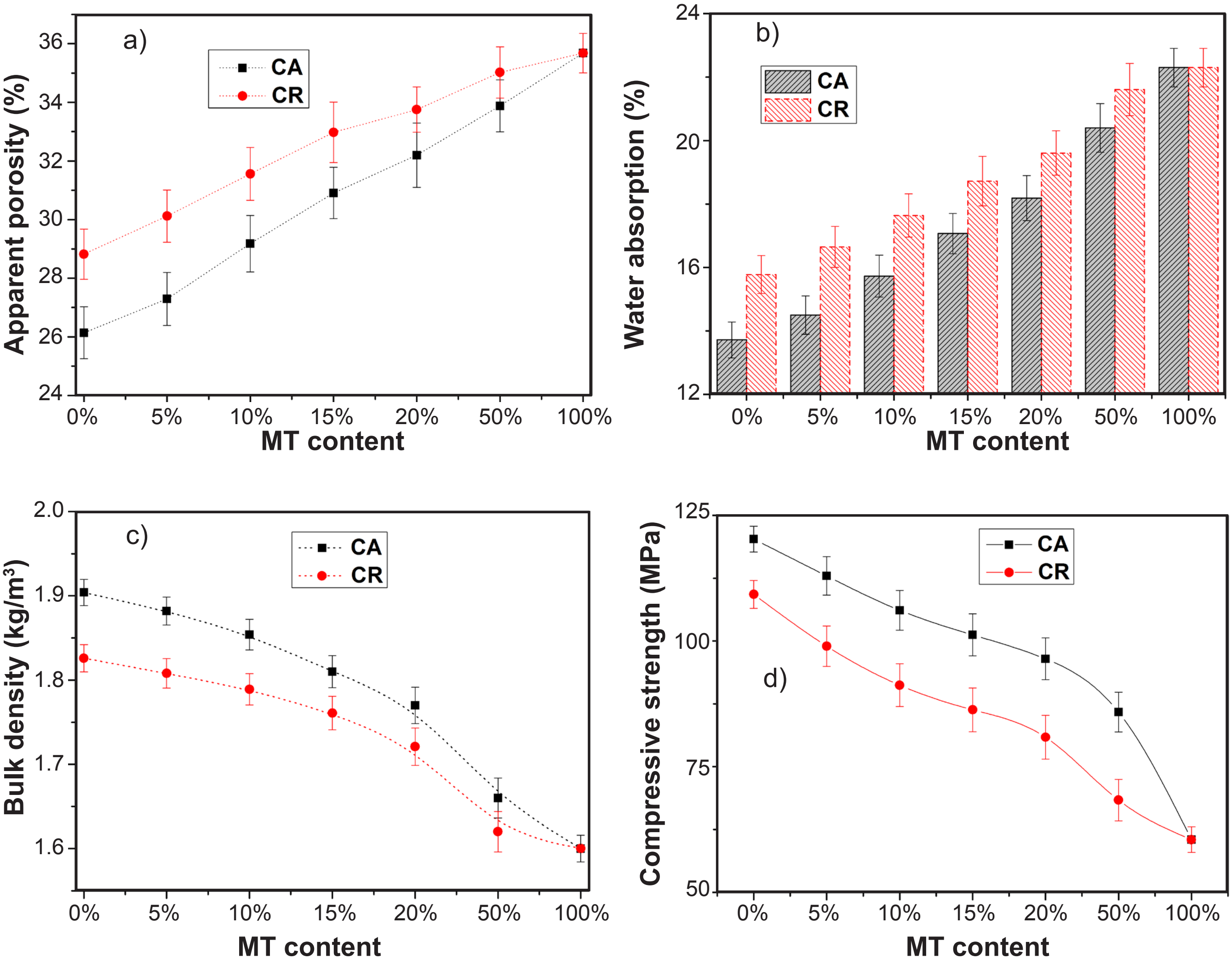

Figure 3: Results of fired bricks' characterization according to the fraction of MT: a) apparent porosity; b) water absorption; c) bulk density; and d) compressive strength.

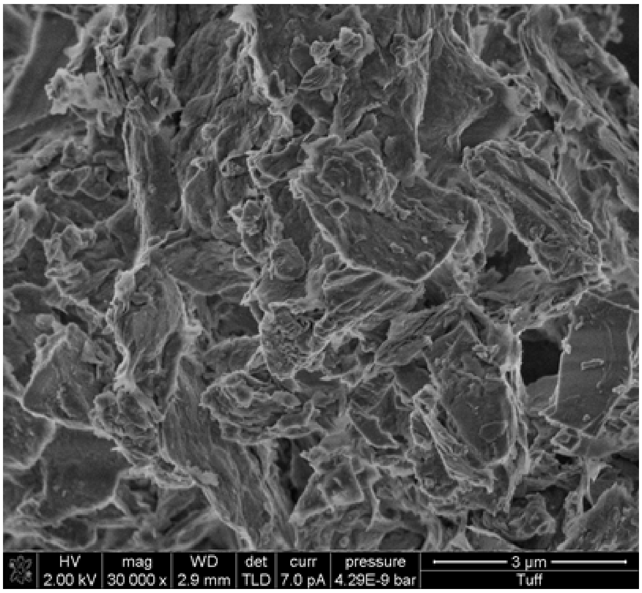

Figure 4: SEM image of MT.

Bulk density: the bulk density of the brick decreased as the percentage of clay substitution by MT increased (Fig. 3c). This could be caused by fewer melting and fusing particles that could thicken brick matrix materials, porosity created during the manufacturing and sintering of clay bricks that prevented microstructural consolidation, and MT's lower real density. The replacement of clay by MT resulted in a coarser particle size in the mixture. The size of the particles is an important parameter since brick produced with a large fraction of sand tends to be more porous, less dense, and more permeable [28].

Compressive strength: as shown in Fig. 3d, the mechanical strength was influenced by the clay substitution rate by MT in the brick. Therefore, increasing the proportion of MT led to a drop in mechanical strength. This decrease ranged from 120 and 109.3 MPa for the reference bricks (without MT) to a minimum of 60.5 MPa for the bricks with 100% of MT replacement. This decrease was linked to the porosity. Higher porosity and characteristics of the pores have significant consequences for the mechanical resistance of the product [29]. At 900 °C, the temperature failed to achieve the melting

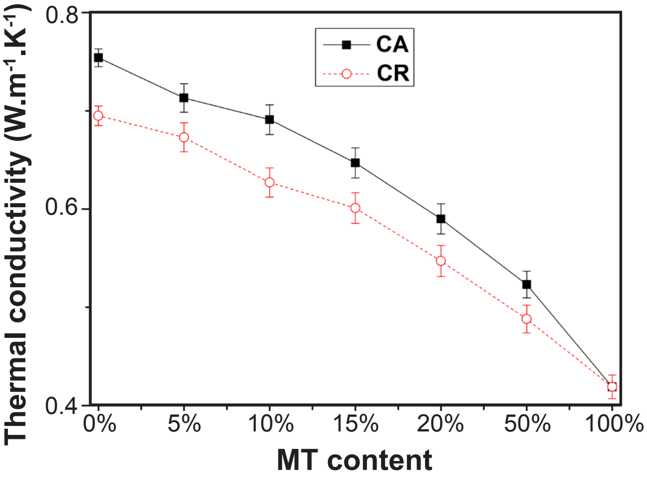

Figure 5: Thermal conductivity of samples according to the fraction of MT.

point; therefore, the bonding between the particles was poor [30]. The deterioration of the plastic properties of the paste makes it less cohesive and more porous, which influences the mechanical properties of the final product and makes it less resistant [31]. The larger particle sizes lead to incomplete sintering, on the other hand, finer ones provide better sintering and more strength [32]. Some of the previous studies [27, 33] reported similar behavior. All values of compressive strength found were still higher than those required by the standards NA 5023 [34] and UNE-EN 772-1 [35] which establish a minimum value of 10 MPa.

Thermal conductivity: materials with lower thermal conductivities are better insulators, reducing the rate of heat transfer to or from the buildings. Therefore, it is important to obtain bricks with good thermal properties. Indeed, the thermal conductivity of regular red bricks is 0.6 $W.m^{-1}.K^{-1}$, whereas insulating bricks have a value of 0.15 $W.m^{-1}.K^{-1}$ [36]. Fig. 5 depicts the thermal conductivity of bricks made with varying percentages of MT. The thermal conductivity of CA and CR control bricks fired at 900 °C was 0.754 and 0.695 $W.m^{-1}.K^{-1}$, respectively. Thermal conductivity decreased when clay was replaced by MT, with a value of 0.419 $W.m^{-1}.K^{-1}$ obtained when clay was replaced with 100% MT. By reducing the bulk density and pore formation, a lower firing temperature allowed to reduce the thermal conductivity [29]. In general, the decrease in thermal conductivity of the specimens with the substitution of clay by MT can be attributed to the increase in the porosity of fired clay bricks. The small amount of fluxing agents in MT to melt and form enough liquid phase might be the cause. Indeed, the short firing time and low firing temperature used during the brick manufacture did not tend the MT to melt. Instead, it was kept in its original state, which improved the brick's thermal resistance and porosity as well as its physical and mechanical properties.

CONCLUSIONS

The construction method and materials selection have a decisive impact on energy efficiency and environmental protection. This work was entrusted to the generalization of a new material based on the mixture of clay and mordenite-rich tuff (MT) for use in the field of thermal insulation in buildings using the ASTM standard. This experimental investigation yielded the following results about the use of MT as a clay substitute in the manufacture of fired bricks for buildings. According to the XRF and XRD analyses, silica and alumina were plentiful in the tuff, which was mainly composed of mordenite. The partial replacement of clay with MT increased open porosity, which caused an increase in water absorption and a decrease in the bulk density of the brick specimens. The thermal insulation qualities may be enhanced by this approach. The compressive strength of the fired samples decreased when the MT was added. The compressive strength of fired clay bricks containing 50% MT decreased by 36.4% and 31.9% using CA and CR, respectively. Finally, the main objective of this study was satisfactorily achieved. Replacing clay with up to 100% MT in brick production can result in better-insulating building material. Thus, the use of MT in large-scale brick production is not only useful for reducing environmental impacts (lower CO_2 emissions) but also for preserving limited clay deposits.

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