



## Research article

## Leaching and geochemical behavior of fired bricks containing coal wastes



Yassine Taha<sup>a, b, \*</sup>, Mostafa Benzaazoua<sup>b</sup>, Mohamed Edahbi<sup>b</sup>, Mohammed Mansori<sup>c</sup>, Rachid Hakkou<sup>a, c</sup>

<sup>a</sup> Mohammed VI Polytechnic University, Materials Science and Nano-engineering Department, Lot 660, Hay Moulay Rachid, 43150, Ben Guerir, Morocco

<sup>b</sup> Université du Québec en Abitibi-Témiscamingue (UQAT), 445 boul de l'Université, Rouyn-Noranda, J9X 5E4, QC, Canada

<sup>c</sup> Université Cadi Ayyad (UCA)/Laboratoire de Chimie des Matériaux et de l'Environnement, Avenue A. Khattabi, BP549, 40000, Marrakech, Morocco

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## ABSTRACT

High amounts of mine wastes are continuously produced by the mining industry all over the world. Recycling possibility of some wastes in fired brick making has been investigated and showed promising results. However, little attention is given to the leaching behavior of mine wastes based fired bricks. The objective of this paper is to evaluate the geochemical behavior of fired bricks containing different types of coal wastes. The leachates were analyzed for their concentration of As, Ba, Cd, Co, Cr, Cu, Mo, Ni, Pb, Zn and sulfates using different leaching tests; namely Tank Leaching tests (NEN 7375), Toxicity Characteristic Leaching Procedure (TCLP) and pH dependence test (EPA, 1313). The results showed that the release of constituents of potential interest was highly reduced after thermal treatment and were immobilized within the glassy matrix of the fired bricks. Moreover, it was also highlighted that the final pH of all fired samples changed and stabilized around 8–8.5 when the initial pH of leaching solution was in the range 2.5–11.5. The release of heavy metals and metalloids (As) tended to decrease with the increase of pH from acidic to alkaline solutions while Mo displayed a different trend.

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## 1. Introduction

The mining industry around the world is criticized for the generation of large amount of wastes with a potentially negative environmental impact. Mining industry, whether underground or open pit, generates three types of solid waste; (i) waste rocks in the form of coarse fragments representing the non-commercial portion of the removed rock; They are generally stored in waste rock dumps, (ii) tailings composed mainly of gangue minerals, water and sometimes chemical additives used during the ore processing phase and finally (iii) contaminated water treatment sludges (Aubertin et al., 2002).

In the context of sustainable development and industrial ecology, several industrial wastes are increasingly recycled as building materials aggregates. The production of fired bricks from alternative materials represents a sustainable way to better manage

tremendous amount of mineral wastes produced by various industries. Several studies investigating the qualities and sources of industrial wastes used in fired bricks making are available in the litterature (Muñoz Velasco et al., 2014; Zhang, 2013; Phonphuak and Chindaprasirt, 2015). A waste classification according to their properties and origin was carried out according to the European waste catalog (EWC, 2002). The amount of wastes that could be incorporated in the fired bricks matrix depends on their physical, chemical and mineralogical properties. Some wastes are used as; fluxing agents (glass wastes, ashes, sludge, etc.), fillers (sandy wastes, ashes, dust, etc.), pore formers (high organic matter and carbonate rich wastes), body fuel agents (high carbon content wastes, oily residues, etc.) and finally as clay substitutes (municipal solid wastes, incineration slag, etc.) (Taha, 2016; Coronado, 2014a; Taha et al., 2016a; Taha et al., 2016b). However, many social and regulation challenges are facing the possibility of recycling potential toxic waste materials in the construction sector. This is due to a lack of knowledge on toxicity and heavy metals release from wastes based bricks. To the best of our knowledge, only few studies has been carried out to understand the environmental behavior of bricks incorporating waste materials (Ukwatta and Mohajerani,

\* Corresponding author. Mohammed VI Polytechnic University, Materials Science and Nano-engineering Department, Lot 660, Hay Moulay Rachid, 43150, Ben Guerir, Morocco.

E-mail address: [yassine.taha@um6p.ma](mailto:yassine.taha@um6p.ma) (Y. Taha).

2017; González-Corrochano et al., 2012). Few leaching tests were used in these studies and the geochemical behavior of contaminants was weakly developed. In contact with water (e.g. rain), potential toxic substances, classified as harmful to the environment, could be released from by-products based bricks. The release of constituents of potential concern (COPC) is mainly affected by the chemical processes and mass transfer mechanisms (diffusion, sorption/desorption, etc.). Their release depends on the physical and chemical properties of the solid material, the nature of the contacting liquid (pH, conductivity, redox potential) and the solid/liquid ratio (Garrabrants et al., 2010; Coussy et al., 2011).

The environmental behavior of constructions products containing primary and/or alternative materials is assessed using different standardized leaching tests. Some tests are based on the Dutch leaching test (NEN-7375, 2004) which is directly coupled to environmental criteria in the Soil Quality Decree (successor of the Building Materials Decree) (SQD, 2007). At the European level, these tests (with minor adjustments) are standardized under the mandate for the Construction Products Regulation (CEN/TS-16637-2, 2014; CEN/TS-16637-3, 2016). In addition, these tests have been the basis for standardization in the US and have led also to very similar protocols. The US is currently assessing the incorporation of these tests in national regulations (SW-846). Various leaching tests are used in the literature depending on their country standards. The most widely used tests can be classified in two categories. The first category consists of the evaluation of COPC released from bloc/monolithic products using diffusion leaching tests (also called tank leaching tests) such as the Dutch test (NEN-7375, 2004). The second category consists of the assessment of the release of contaminants from ground wastes based bricks such as the American test; Toxicity Characteristic Leaching Procedure (TCLP) (US-EPA, 2009) and the European test; Compliance Leaching Tests for Granular Materials (CEN/EN-12457, 2003). Another type consists at studying the leaching behavior of COPC from granular wastes subjected to different pH controlled solutions (pH between 2 and 14); known as pH dependence tests (Method-1313, 2012). The results of each test in terms of leachate quality are usually compared with the thresholds of elemental concentrations, established by the respective standards.

The current study is the logical concomitant of the previous study (Taha et al., 2017) where the physical and mechanical properties of bricks produced from both natural and coal mining waste materials were investigated. The basic idea of this paper is to contribute to the knowledge improvement on environmental behavior of mining wastes based bricks. The goal is to assess the effect of thermal treatment on the leaching quality of fired bricks containing coal wastes. Two types of coal wastes were evaluated: coal waste rock (CMWR) and corresponding treated coal tailings (TCMT). A reference brick containing only natural shales (ShB) is used as a reference material. The leaching tests conducted in this study are realized on both raw materials (CMWR, TCMT and ShB) and fired bricks.

## 2. Materials and methods

### 2.1. Raw materials

Three raw samples were used in this study to manufacture cylindrical brick samples. A reference material consisting of shales for bricks (ShB) and two types of coal wastes; coal waste rocks (CMWR) and corresponding treated coal tailings (TCMT). TCMT is the result of the decarbonation of CMWR by froth flotation process (Taha et al., 2017). Coal (to be valorized) was recovered from CMWR and the resulting tailings TCMT was used to manufacture good

quality bricks. A representative sample of CMWR was collected from the coal dumps of Jerada city in Morocco. This site contains more than 20 million tons of coal wastes. These wastes represent various risks related to its chemical, physical and mechanical instability.

### 2.2. Physical, chemical and mineralogical characterization

The raw materials were sieved at 2 mm, dried, homogenized and preserved in sealable plastic bags before testing. The grain size distribution was performed using a laser analyzer (Malvern Mastersizer). The specific gravity (Gs) was measured with a helium gas pycnometer (Micromeritics Accupyc 1330). The major elements were analyzed using an X-ray Fluorescence Analyzer (Bruker, Tiger Model). The trace elements were determined after a multi-acid digestion (HNO<sub>3</sub>/Br<sub>2</sub>/HF/HCl) followed by Inductively Coupled Plasma with Atomic Emission Spectroscopy (ICP-AES, Perkin Elmer Optima 3100 RL) analysis. The crystalline phases present in the wastes and in shales were determined by X-Ray diffraction (Bruker AXS Advance D8, Cu-K $\alpha$  radiation). The DiffracPlus EVA software was used to identify mineral species and TOPAS software to quantify the abundance of all identified minerals.

### 2.3. Fired bricks manufacturing

CMWR and TCMT were investigated for their recycling in manufacturing fired bricks (Taha et al., 2017). The results of this study showed that bricks of good quality could be manufactured from only CMWR and TCMT samples. The previously optimized formulations and processes were used. The mixtures of raw materials and water were homogenized, mixed and pressed under 6 MPa using a hydraulic press to manufacture cylindrical blocks measuring 5.4 mm of diameter and 38 mm of height. The unfired samples (green samples) were then dried at room temperature during 24 h followed by a controlled drying process in an electric oven at 60 °C during 24 h. Once dried, the bricks were fired in a muffle furnace (Nabertherm<sup>®</sup>) with a ramp rate of 30 °C/h and held at the optimal temperature 1020 °C during 5 h.

### 2.4. Physical and mechanical characterization of fired bricks

The mechanical strength was investigated using a universal testing machine (Zwick Roell) with a load capacity of 30 kN, according to the ASTM-C67 (ASTM-C67, 2003) standard. The apparent density, water absorption, and apparent porosity of fired bricks were measured according to ASTM-C373 (ASTM-C373, 1999). The firing shrinkage was measured according to ASTM-C326 (ASTM-C326, 2003).

### 2.5. Leaching experiment setup

The experimental approach followed in this study is described in Fig. 1. The leaching behavior of raw materials and fired grinded bricks was assessed according to Toxicity Characteristic Leaching Procedure (TCLP) (EPA-1311, 1992) and results are compared with US-EPA (US-EPA, 2009) thresholds. It is aimed to evaluate the effect of firing on the mobilization of COPC. Furthermore, different categories of tests were used to evaluate the leaching behavior of fired bricks using; Tank Leaching Test (NEN-7375, 2004) on monolithic bricks and pH dependence test (Method-1313, 2012) on granular bricks (grinded bricks).

#### 2.5.1. Toxicity characteristic leaching procedure (TCLP)

The raw materials were tested in their initial form while fired

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