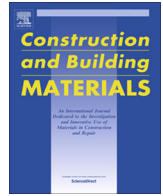




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## Review

# Processing, effect and reactivity assessment of artificial pozzolans obtained from clays and clay wastes: A review



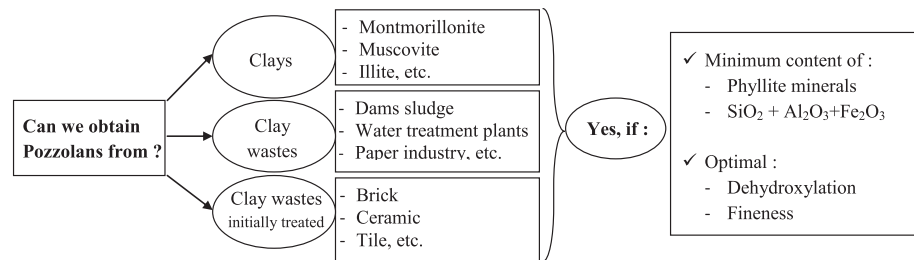
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### HIGHLIGHTS

- Clays, clays wastes and clay wastes initially treated can lead to pozzolans.
- In these materials, phyllite minerals are at the origin of this pozzolanicity.
- To elaborate an APC, the treated material must be totally dehydroxylated.
- Clay wastes mineralogical variety make difficult to elaborate a high reactive APCW.
- In the APCWIT case, the initial treatment uncertainty is the major disadvantage.

### GRAPHICAL ABSTRACT



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### ABSTRACT

The blended Portland cement, CEM II, is obtained by replacing a part of clinker by an addition, as described in the EN 197-1 standard. Among the materials cited in this standard, there are the artificial pozzolans. When used as partial replacement of clinker or cement and in presence of water, the pozzolanic materials react with the calcium hydroxide  $\text{Ca}(\text{OH})_2$ . This reaction leads to a Supplementary Cementitious Compounds SCC (C-S-H, CAH, CASH) comparable to those formed during the ordinary cement hydration. Treated clays are an artificial pozzolans widely studied in the last years. It is known that these materials are essentially composed from phyllosilicates, quartz, carbonates, etc. As the pozzolanic reactivity depends mainly of phyllite minerals, several features must be taken into consideration, especially the dehydroxylation rate. Generally, the dehydroxylation is obtained by thermal treatment, which varies from clay to another. Recent studies have shown that the use of treated clays may lead to improvements in mortars and concretes properties, whether at the fresh state or the hardened one. The assessment of these improvements is often conducted by many techniques and tests allowing to estimate the treated clays pozzolanicity. This paper presents an overview of the literature related to the elaboration, the utilization, the efficiency and the pozzolanicity tests of some clay minerals and wastes used as reactive additions in blended cements.

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## 1. Introduction

The Portland cement is a powdery substance made from some naturally occurring minerals, mainly contained in limestone and clay. In this mix, CaO is the major component, while oxides: SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub> and Fe<sub>2</sub>O<sub>3</sub> are supplied by the clay, initially used at about 20%. After grinding, the raw meal is burned at around 1450 °C, where, chemical reactions take place to form the clinker, which contains, essentially, calcium silicates (C<sub>3</sub>S, C<sub>2</sub>S) and aluminates (C<sub>3</sub>A, C<sub>4</sub>AF). During the cement production process, CO<sub>2</sub> is emitted by two different sources: the CaCO<sub>3</sub> decomposition (decarbonation) and the combustion of fossil fuel (during the burning). Cement manufacturing is considered to be one of the highest carbon dioxide emitting industries in the world by emitting almost 0.83 kg of CO<sub>2</sub> per each kg of cement produced [1]. This industry generates approximately 5% of anthropogenic CO<sub>2</sub> in the world [2] and can reach 8%, according to Kajaste [3]. Among the solutions proposed to reduce this level of CO<sub>2</sub> emissions from cement production, we cite: the use of blended Portland cement, in which we proceed to the partial substitution of clinker by some additions.

Pozzolanic additions are widely studied in the last decades. These supplementary cementitious materials (SCMs) may be used as a substitution of clinker [4,5], or cement [6]. Whatever natural [7] or artificial [8], these materials are used to improve some characteristics of mortars as strength [9], durability [6], rheological and transfer properties [10]. The enhancement of these properties is linked to the reactivity of these additions leading to a Supplementary Cementitious Compounds SCC (chemical pozzolanicity) [11], but also to their fineness (physical pozzolanicity) [12].

As artificial pozzolanic additions there are two types: by-products such as fly ash and silica fume [6] and treated clays where the most known is Metakaolin. The Metakaolin is obtained from a kaolinitic clay after heat treatment between 500 and 900 °C. It has attracted the interest of several researchers [13–15] and was the subject of the NF P-18 513 standard. Some other clays may lead to artificial pozzolans, they must be subjected to treatment (thermal in usually), ensuring a maximum of dehydroxylation, without recrystallization. This is the case of *illite* [16,17], *smectite* [18,19], *bentonite* [20], *muscovite* [21], *montmorillonite* [16,22], *chlorite* [23], *sepiolite* [24], low-purity mica clay [25], etc. Besides the exploitation of natural resources, recent studies have shown that it is possible to obtain artificial pozzolans from some sludges (after treatment) such as: dams [26,27], water treatment plants [28], paper industry [29], and wastes (without treatment) such as: brick, tile, marble and ceramic industries [30–32]. These materials have received lesser attention, probably because of their lower reactivity and subsequently lower performance as SCMs in cement blends [19].

It is known that the clays are mixtures of clay minerals (phyllosilicates) and clay crystals of other passive phases such as quartz,

carbonates, feldspars and metal oxides. Phyllosilicates are a large family of minerals that commonly show layered structures [33]. Phyllosilicates, mostly composed of silica and alumina, present a sharp pozzolanic activity when calcined and ground to cement-fine [34]. If they are well thermally-treated, these minerals lose their crystallinity, and became able to release reactive silica and alumina leading to SCC production by consumption of Portlandite. To understand artificial pozzolans elaborated from clays, knowledge of clay minerals is indispensable.

Clay minerals may be classified according to their layered structure as types 1:1 or 2:1 (Fig. 1). Each layer is fundamentally built of one or two tetrahedral silicate (Si-O) sheets and one octahedral metal (Mg<sup>2+</sup> or Al<sup>3+</sup>) – oxide or hydroxide (M-O or M-OH) sheet. The 1:1-class clay mineral consists of one tetrahedral sheet and one octahedral sheet, like kaolinite and serpentine. However, the 2:1-class is composed of an octahedral sheet sandwiched between two tetrahedral silicate sheets, like vermiculite, montmorillonite and sepiolite [33]. Some atoms (Si, Mg and Al) can be substituted for other atoms, which induce negative charges all over the layers. As a result of these negative charges, interlayer cations (K<sup>+</sup>, Ca<sup>2+</sup>, Na<sup>+</sup>, etc.) must be absorbed.

Clay wastes can be classified into two categories, (i): Clay wastes (CW) requiring a treatment, like marine sediments, sludge of dams, water treatment plants, etc. (ii) clay wastes initially heat treated (CWIT), we cite: wastes of brick, ceramic or tile.

At the raw state, the clay has two kinds of water: physically bound (adsorbed) and chemically bound which takes the form of hydroxyls in the clay phyllosilicate minerals. The efficiency of artificial pozzolans obtained from clays (APC) or from clay wastes (APCW) depends on the removal level, by treatment, of the chemically bound water (dehydroxylation). The total dehydroxylation corresponds to the destruction of these minerals crystallinity, which confers them an amorphous state and a pozzolanic reactivity. However, the reactivity of artificial pozzolans obtained from clay wastes initially treated (APCWIT) is difficult to be controlled, due to their inappropriate treatments.

The use of clays and clay wastes, as pozzolanic additions in cement industry, is well known throughout the world. While (APCW) and (APCWIT) are still under study and not yet normalized or commercialized, the industry of (APC) is widely developed especially for Metakaolin. In France, since 1983 the annual production of MK is 200,000 tonnes. The material, commercialized under the NF P18-513 standard, has already been used in many projects, including the bridge Chaban-Delmas in Bordeaux. However, the world's total output of kaolin exceeds 25 million tonnes [35]. According to Mirovic et al. [8], a large kaolin clay deposit of about 55 million tonnes, is located in "Garaši" basin in Serbia and not yet exploited. Haili Cheng [36] reported that more than 20 billion m<sup>3</sup> of clay brick products, produced in China in the past five decades, will mostly be transformed into clay wastes in the next five

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