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Synergistic recycling of calcined clayey sediments and water potabilization sludge as geopolymer precursors: Upscaling from binders to precast paving cement-free bricks

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HIGHLIGHTS

- Calcined sediments and potabilization sludge were used as geopolymer precursors.
- Precast geopolymer paving bricks were produced after upscaling process.
- Significant results were obtained for sustainable production of building materials.
- The proposed recycling route is very advantageous for reservoir management.

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ABSTRACT

The building materials industry is facing relevant challenges in terms of sustainability requirements. The same challenge is expected for other anthropogenic activities such as reservoir management. In this study, in the light of industrial ecology approach, two wastes, namely clayey sediments and water potabilization sludge, generated through reservoir life cycle, were used in a synergistic way in the synthesis of sustainable geopolymer binders. In order to guarantee a productive equilibrium between the different yearly evolution of building materials demand and wastes production by the basin, precast materials have been regarded as optimum potential application. In this regard, calcination conditions, mix design and curing conditions were preliminarily optimized. Particularly, geopolymerization kinetics were evaluated by means of mechanical and microstructural characterization of pastes to assess the influence of early age curing conditions and mix design on the engineering performance and, afterwards, the product was upscaled to a typical precast concrete element. The whole set of results demonstrated the feasibility of the proposed recycling route, revealing highly promising perspectives for further studies and broader application field.

1. Introduction

Recent updates in legislative approach in reservoir water and sediment management revealed the need to make supplementary efforts in research towards the definition of possible reuses of produced wastes [1–3]. Due to its high production volumes and simultaneous need of increasing sustainability, building materials industry represents a highly promising solution to recycle the total volume of sediments produced by silting in a reservoir. The use of harbor, river and reservoir sediments was proposed in brick production [4–6], sintered lightweight aggregate manufacturing [7–12] and production of Portland cement [13]. Hence, summarizing the main results of the mentioned literature dealing with the beneficial reuse of sediments, two main application-oriented research trends can be detected, namely bricks and sintered lightweight aggregates. In both cases, high temperature processes are required, implying a harmful environmental effect. In the latter case, more sustainable lightweight aggregates can be obtained with alternative low temperature processes, such as cold bonding pelletization [14–16]. In the case of bricks, and, generally, precast elements, firing can be substituted by optimized calcination treatment of clayey sources and proper mix design. The role of

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calcination is crucial in the development of proper performance of clay-based binders, and this was clarified since the first pioneering works [17–21], where calcined clays were employed as pozzolanic admixture in cementitious systems. One of the main problem concerning the recovery of sediments is represented by the heterogeneity of clay sources, as highlighted in Table 1. As it can be observed in this table, the structural performance can vary in a very wide range due to different chemistry and reactivity, going from load-bearing elements (laterite/pure kaolinite) down to non-structural materials (nature impure clays/Illite/etc.). Starting from this consideration, it is clear that a proper and economically feasible recycling track for dredged sediments cannot be associated a priori to high performance structural materials. In this scenario, the production of bricks and other precast non-structural elements can be proposed with higher reliability and elasticity respect to periodical oscillations in raw material availability. Another main technological issue is represented by calcination parameters. Three main calcination equipment can be found nowadays in clay processing, with different operating conditions: rotary furnace, multiple earth furnace and flash calcination plants. The latter one gained a lot of research attention due to increased sustainability in terms of energy demand [22] and higher efficiency in particle treatment. In the work by San Nicolas et al. [23], a description of the process is provided. After a preliminary drying process, the clay-based raw mix is crushed and coarse particles and quartz are eliminated. A secondary drying process is then performed at 100 °C. Afterwards, a stream of hot air takes clays to 200 μm sieving just before feeding. The calcination phase is carried out for few tenths of seconds of 1000–1200 °C. The process ends with air cooling cyclones and material storage in silos.

The same study provides an evaluation of influence on rheological properties is carried out by producing mortars with paste composed by cement and different metakaolin sources with fractions equal to 75 and 25%, respectively. The rheological measurements used was LCL flow test (EN 196-1) and the reduction in flow time due to provided by flash calcination for different clay source was in the range 38.0–62.5% [23]. These results contribute to clarify the need of optimizing calcination process.

Alumina-containing water potabilization sludge (WPS) is another fundamental residue produced by reservoir management activities. This waste is accumulated after treatment processes for water potabilization, which are based on flocculation-clarification using alumina-based coagulant [24]. The amount of generated sludge and its chemical composition mainly depend on the following parameters: (i) water chemical and physical characteristics; (ii) efficiency of removal process; (iii) type and dose of coagulant. The amount of sludge can be approximately estimated to be in the range 1–5% of the total untreated water quantity [24]. This waste was investigated in literature only in few studies, mainly concerning the potential reuse in building materials industry. Particularly, WPS was proposed in the production of bricks [25–27], ceramics [28,29], cement manufacturing with role of alternative raw material [30–34], inorganic binder-based composites [35–40], lightweight aggregates [41,42]. Furthermore, WPS were considered in alternative civil engineering applications such as soil improvement and other geotechnical works [43]. In a recent study [44], the pozzolanic potential of calcined water potabilization sludge (CWPS) was investigated in a deeper way than preliminary literature studies. In this work, Chapelle test was applied by preparing cementitious blends with lime and CWPS (alumina content equal to 16.47% according to XRF results) as pozzolan, after a calcination process carried out at 400–900 °C for 2 h. The highest amounts of fixed lime were found at 700 and 800 °C, with a detrimental effect of further temperature increase associated with the initiation of crystallization processes of amorphous silica [44].

The use of CWPS and calcined clay sediments (CCS) can only partially reduce the environmental impact of cement, mainly associated with the content of clinker, whose production is responsible on a global scale for 5–8% CO_2 emissions. Hence, in order to further increase the sustainability of the recycling process of both CWPS and CCS, cementitious blends should be avoided. In this framework, the use of alternative low-energy and low-CO_2 binders such geopolymers (also called "alkali activated binders" or, more recently, "hybrid cements") is highly promising. Geopolymers were proposed for a wide range of engineering applications such as: production of geopolymer concrete [45,46]; production of bricks and other non-structural elements starting from "poor" solid precursors [47,48]; waste stabilization/solidification [49,50]; fire resistant binders [51,52]; and other functional applications [53,54].

The idea of finding a common solution for sustainability issues of two fundamental anthropic activities such as reservoir management and building materials production through the geopolymerization of CCS was already proposed in literature [55–57]. In [55], a preliminary investigation proved that sediments from two Italian reservoirs located in Southern Italy had the potential to be employed as aluminosilicate solid precursors for the synthesis of geopolymers after calcination at 650 and 750 °C, considering that sufficient mechanical performance for non-structural components was already achieved. In a subsequent study [56], FTIR and NMR analysis confirmed that, in the range 400–750 °C, the dissolution of both Al and Si increased with temperature, revealing also that the addition of blast furnace slag as secondary component of the blend can significantly increase the reactivity. More recently, Peirce et al. [57] considered the possibility to use an aluminate alkaline solution to activated CCS in place of typical silicate solutions, achieving good mechanical performance.

In this study, for the first time, the synergistic recycling of CCS and CWPS to produce precast geopolymer concrete elements was proposed. Particularly, starting from characterization of clayey sediments and water potabilization sludge, the effect of calcination was evaluated and an evaluation of optimal sediment/sludge ratio was provided by testing geopolymer pastes from a mechanical and microstructural point of view. Afterwards, a gradual manufacturing upscaling from paste to mortars up to real scale elements (paving bricks) was carried out, and main rheological and mechanical issues were highlighted. Significant insight into several key parameters were obtained, particularly mix design, calcination parameters, workability, curing conditions, mechanical properties, were

Table 1
Data collection related to mechanical strength variations due to heterogeneities in clay sources (adapted from [21]).

Clay source	Mineralogical description	Compressive strength [MPa]	Strength loss
Laterite	Kaolinite (55%); quartz; iron oxides	27.1	/
Pure kaolinite	Kaolinite (>98%); quartz (<2%)	26.9	−0.74%
Tropical soils	Kaolinite (33 ÷ 44%)	15–23	−15.1 ÷ 44.6%
Montmorillonite	Montmorillonite (>90%); quartz	12.6	−53.5%
Natural impure clays	Kaolinite (30%); illite (30%); quartz	8–9	−66.8 ÷ 70.5%
Illite	Illite (>85%); quartz	7.8	−71.2%
Slate waste	Chlorite (29%); muscovite (46%); quartz (24%)	2.1	−92.2%
Black colliery spoil	Kaolinite + Illite/mixed layer clay; quartz	0.6	−97.8%