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Ceramic light-weight aggregates production from petrochemical wastes and carbonates ($NaHCO_3$ and $CaCO_3$) as expansion agents

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HIGHLIGHTS

- A novel material was produced from petrochemical wastes.
- This material features light-weight aggregate properties.
- A particle density of 1.223 g/cm^3 was achieved from the mixture CW_$NaHCO_3$_40 sintered at 900 °C.
- The resulting material exhibits a foam-like structure with rounded cavities.

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ABSTRACT

This work shows the production of a ceramic light-weight aggregate (LWA) from mixtures of petrochemical wastes (oily wastes and oil sludge), a conventional clay and two different carbonates ($NaHCO_3$ and $CaCO_3$) used as expansion agents. The novelty of this study is the valorization of an important hazardous waste for the synthesis of a potential material for construction. All these feedstock where characterized by elemental, proximate and TG/DTG analyses and also by XRF for determining the inorganic composition. The LWA were produced from two different mixtures: i) clay and oily waste (CW) and ii) clay and oil sludge (CS), by using different proportions of both $NaHCO_3$ and $CaCO_3$ (0, 20 and 40 wt%). Additionally, the influence of sintering temperature (900, 950 and 1000 °C) on the particle density was also evaluated. Although TG/DTG analysis suggests a higher CO_2 release with $CaCO_3$ and hence, a higher bloating phenomena; there also appears an influence of the petrochemical waste composition. In addition, the particle density varied from 1.394 to 2.227 g/cm^3, and from 1.226 to 2.571 g/cm^3, for mixtures using $CaCO_3$ and $NaHCO_3$, respectively, and the lower the sintering temperature, the lower the particle density. The lowest density (1.223 g/cm^3) was obtained when oily waste and $NaHCO_3$ at 40 wt% are used (CW_$NaHCO_3$_40). For this mixture, the synthesis process is prompted not only by the $NaHCO_3$ addition, but also by the reaction among different elements of clay and oily waste. The resulting aggregate exhibited a foam-like structure with rounded cavities, suggesting the bloating phenomenon occurrence.

1. Introduction

The petroleum sludge is a kind of waste generated by the petrochemical industry as consequence of sedimentary hydrocarbon compounds that have been agglomerated by long periods in broad fields [1]. It is generated as result of certain petroleum refineries activities such as fluid catalytic cracking (a processes that uses a catalyst to crack large hydrocarbons molecules into smaller ones, usually known as FCC), and visbreaking (a process that reduces the viscosity of large hydrocarbon molecules by thermal cracking leading to smaller hydrocarbons ones), and also from the wastewater treatment in this industry [2]. Besides heavy hydrocarbons, this sludge is also composed by water, heavy metals and inorganic compounds, among others [3]. Due to these complex characteristics, this sludge has received a special attention and it is currently recognized as a hazardous waste [4,5]. Hence, these facts make no possible to be discharged in landfills, even if they are de-oiled; and its improper or insufficient disposal can cause serious effects to both the environment and the human health [5].

The total petroleum sludge generation in Colombia, including all the types of oily-wastes, was estimated to be around 108,000 tons in 2013 [6]. This amount corresponded to 45% of the total haz-

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ardous wastes generated in this country (≈240,000 ton) [6]. Around 52% of the total oily-wastes generated were mainly treated by physicochemical procedures, while the rest was re-used through processes of recovery, recycling or reclamation (17%) and disposed in a secure landfill/cell (31%). Therefore, almost a third of these wastes faces a serious environmental problem as well as an important feedstock squandering.

On the other hand, there is an increasing trend towards valorization of materials that have ended their useful life, as a way to reduce wastes requiring landfill, in line with the waste management hierarchy [7,8]. In this sense, there are many materials than can be used as fillers for building and/or as direct construction material. In fact, many new materials have been developed from industrial wastes for construction applications [9]. Generally speaking, they are known as aggregates and are classified according to: a) coarse and fine, depending on size; and b) light-weight, normal-weight and heavy-weight, depending on density [10–12]. These aggregates are porous granulated solids that could be natural or synthetic. The former ones can be obtained from different natural sources such as volcanic, sedimentary or metamorphic rocks [12,13]. The latter ones are found mainly from industrial wastes such as fly ashes and other unconventional raw materials such as municipal solid wastes, sediments from dredging, and mining and quarrying residues, among others [14–17]. These aggregates are commonly produced by mixing the aforementioned feedstock with different clays and minerals, and can also involve the use of a cement matrix as a binder [13,18–20].

Among all them, the most frequent aggregate is the lightweight (LWA). According to EN-13055-1 [10] the LWA must have a loose bulk density lower than 1.20 g/cm^3 or a particle density lower than 2.00 g/cm^3. This low-density aggregate is commonly produced starting from minerals, including natural rock materials such as shale, expanded clay, perlite, vermiculite, or slate, and different kinds of pelletized or sintered waste, including in many cases sintered glass [21,22]. These raw materials are thermally expanded to about twice the original volume to get final aggregate density. [22] LWA was first manufactured commercially in the UK during the 1950s using clay and shale from the mining and slate industries [23].

The porosity development and hence, the density of the aggregate is generally explained by the gases released in the total or partial oxidation of the organic matter included in all the feedstock [17]. Similarly, the presence of foaming additives also known as expansion agents, such as Co_2O_3, $SrCO_3$, $CaSO_4$, MnO_2, talc, and water-glass, enables the release of gases within the range between the softening point and the maximum sintering temperature [21]. Therefore, the porosity development and the consequently expansion of the ceramic solid (bloating) takes place. Prior to the gas release, a viscous liquid phase is formed. This pyro-plastic deformation retains bubbles inside the aggregate giving as a result its foaming structure.

Although is hard to identify the critical components that cause bloating [23], it seems that two different but inter-related conditions are essential to form this structure [24]. The first one is associated with the material ability to form a glassy phase with high viscosity at elevated temperatures in order to trap the gases released. The second condition is related to the chemical nature of the raw materials, which must have a certain chemical composition in order to favor the devolatilization process of some compounds at that temperature where the glassy phase is formed. Some examples of this behavior have been reported in the production of LWA from water reservoir sediment [25] and from washing aggregate sludge and fly ash [26]. The limits of bloating have been defined by Riley [24] and Cougny [27] in terms of Al_2O_3, SiO_2 and fluxing compounds ($Fe_2O_3 + Na_2O + K_2O + CaO + MgO$), and Al_2O_3, Fe_2O_3 and alkaline-earths compounds ($MgO + CaO + Na_2O + K_2O$), respectively.

These fluxing compounds not only play an important role for lowering the sintering temperature, but also to promote the bloating phenomena given the occurrence of exothermic reactions. For example, the effect of the temperature over the expansion ability of ceramic products as consequence of the content of CaO and MgO was studied in ceramic glazes formulations [28]. Other studies have reported the effect of amorphous silicon oxides (SiO_2), in form of cullet powder, on the bloating behavior of aggregates made of different raw materials such as fly ash, water reservoir sediment, sand sludge and zeolitic rocks among others [18,29]. In the same way, there are some studies related to the enhancement properties of aggregates by using different additives [7]. All these works show that these inorganic compounds affect the viscosity of the resulting product, promoting the bloating phenomena.

As can be inferred from the above, there are many scientific works on the production of aggregates from several materials including wastes. However, according to the best authors' knowledge, there is no major information on the production of these aggregates from an important hazardous material as petrochemical wastes, and much less on the effect of carbonates over its most important property: density. Therefore, the present study is focused on the production of a ceramic LWA from these petrochemical wastes adding $NaHCO_3$ and $CaCO_3$ as expansion agents to reduce density.

2. Materials and methods

2.1. Raw materials

The raw materials used for the production of aggregates were mainly constituted by clay, (i) an oily waste, found at the bottom of the oil storage tanks; and (ii) an oil sludge, generated in different drilling operations. Both wastes are produced in the petrochemical processing industry and were provided from a Colombian oil company. In addition, sodium bicarbonate ($NaHCO_3$ ~99%) and calcium carbonate ($CaCO_3$ ~99%) were used as expansion agent.

The elemental analysis of both petrochemical wastes was performed with a CHNS Perkin Elmer/O Analyzer 2400. Likewise, the inorganic content (ash) for both petrochemical wastes was determined according to EPA 160.3 SM-2540-G. The inorganic compounds were measured by X-Ray fluorescence (XRF) carried out with a Philips equipment model PW 2400 with a 3 kW tube. This technique was also used for determining the composition of the clay. In addition, different thermogravimetric experiments were conducted for the carbonates, the clay, the petrochemical wastes and their mixtures, by using a thermobalance system Mettler Toledo TGA-SDTA 851. These tests were conducted from room temperature to 900–1000 °C. The sample weight used in all experiments was around 20 mg and the carrier gas was N_2 (99.99%) at 60 mL$_N$/min.

2.2. Aggregate manufacturing

The ceramic aggregates were prepared by using two different mixtures: one composed by clay and oil sludge, herein named CS; and another one denominated CW made of clay and oily waste. Before including the expansion agent ($NaHCO_3$ or $CaCO_3$), an initial mixture was prepared by using 85 wt% of clay and 15 wt% of the petrochemical waste, either oily waste or oil sludge. This blend proportion was obtained after have carried out several tests seeking to avoid a flame occurrence (oxidation) during the sintering process. All raw materials were mixed and conformed manually, without water addition, until a homogenous mass in spherical