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Design of ceramic filters using Clay/Sawdust composites: Effect of pore network on the hydraulic permeability

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ABSTRACT

Clay based ceramic composite materials with hydraulic permeability were elaborated using sawdust as porogent agent. Their mechanical, morphological, microstructural and pore network properties were investigated. Mixtures in various ratios of two kaolinite clay minerals, *Ba* (highly plastic) and *Va* (sand-rich) constitute the five ceramic matrixes studied (CM1, CM2, CM3, CM4 and CM5). Due to their high flexural strength, CM3 and CM4 received 0%, 5%, 10% and 15% sawdust before firing, to improve the porosity of the final matrixes. Results revealed that 900–1000 °C is the range of temperature necessary to get good sintering and flexural strength (≥2 MPa). A typical clay-sawdust based materials (parallepipedic bricks) present porosity ≥40 vol% and 1.5 g/cm^3 density. Characterizations such as FTIR, SEM, MIP and flow permeability of ceramic candles were performed. A Hydraulic permeability of ~10 mDarcy was obtained and the mean pore diameter varies from 0.05 to 0.1 μm, in agreement with the microstructure exhibited by the ceramic candles. In the presence of sawdust, pores with size up to 10 μm were observed, justifying the increase of flowing permeability. The elaborated matrixes are promising candidates for microfiltration.

1. Introduction

A recent United Nations (UN) report on enhancement of resources in water reveals that 663 billion people lack access to sources of potable drinking water and 2.4 billion live without adequate sanitation services [1,2]. Africa possesses the weakest infrastructure and equipment for cleaning up. Yet, 36% of the African population lack good sources of water and 70% do not have adequate sanitation services. Even where the provision of drinking water is shown to be satisfactory, the question of treatment/purification of wastewater remains problematic. In fact, in Africa and in most developing countries, about 90% wastewater is rejected in surrounding environment without treatment [3,4]. This situation leads to many consequences: cholera and water bone diseases affect thousands of people worldwide. Developing countries do not have enough water purification system and the few part made from activated sludge suffer from maintenance [5] or from climate accommodation [6]. Besides, the efficiency of ceramic devices in removing various pollutants, bacteria elimination notably has been clearly demonstrated [7–9]. The purification of wastewater by using porous ceramic based on waste resources has been documented [10]. As shown by many authors, the porosity and hydraulic permeability of some ceramic filters are improved by waste resources such as sawdust, rice and coffee husks [11,12].

Porosity is known as the most meaningful property to describe a porous material as it allows to indicate the volume of emptiness (cavity). Porosity is expressed as the fraction of total volume taken up by pore space. Its quantification is essential to obtain parameters such as thermal conductivity, mass transfer, diffusion coefficient and permeability [13]. For a material, the permeability (k) is the ability to let itself cross by a fluid under the effect of a pressure gradient. This constant is used to evaluate flowing of liquid through a porous medium and is expressed in Darcy. The ease at which water flows through a porous ceramic corresponds to the level of permeability [12]. The types of pores in porous materials that contribute to the processes of filtration are isolated and interconnected pores [14]. However, the composition of the ceramic matrix, as well as heating are factors that influence the porosity [15,16]. It has been reported that the microstructure of a ceramic filter present small particles sizes ranging from

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0.02 to 4 μm and that their porosity can vary from 34% to 46% [9,17]. However, an additional pore from the action of porogent agent in ceramic filter can contribute to increase the flow rate. There is also a vesicular structure presenting interconnected round pores of 3–4 μm forming the ideal channels for the hydraulic conduction [9]. So at a given particle size distribution, pore radii and volumetric porosity correspond a precise permeability which can decrease as grain radii decrease. Permeability is therefore a function of porosity, grain size, packing arrangement (size and pores), capillary structure, pore connectivity and orientation [18,19]. The hydraulic conductivity (cm/s) can be high ($k > 10^{-1}$), median ($10^{-3} < k < 10^{-5}$) or low ($10^{-3} < k < 10^{-7}$) [9]. This corresponds to the cases of industrial wastewater, simple wastewater and drinking water respectively. The hydraulic retention rate of ceramic filter depends on the shape of the filter and can range from 5 to 10 L/day according to literature [7,9,12].

In spite of the abundance of clay minerals in Africa, very few research has been carried out on the production of filter materials based on these low-cost and available materials. Recently, Belibi et al. [20] have exploited some cameroonian clay minerals to elaborate support for microfiltration. According to the index of the clay deposits in Cameroon, the Southern part is endowed with a lot of kaolinite. For instance clay from Center region in Mvan deposit is used to produce fired refractory bricks [21]. Clay minerals from Bamessing (Nord-West region, Cameroon) are used locally for traditional pottery [22]. To obtain the desired properties such as color, plasticity or mechanical strength, the Local Material Promotion Authority (MIPROMALO, Yaounde, Cameroon) uses mixtures of clay minerals from Bamessing and Mvan (Center region, Cameroon) to produce decorative ceramic objects. To the best of our knowledge, hydraulic and microstructural properties for filtration of fired clay from these deposits have not been studied. It is therefore necessary to explore filtering capacity of these clayey systems reinforced with sawdust to produce water purification devices. This work aims to produce ceramic devices from a mixture of clay minerals with sawdust and to evaluate their physic-chemical, mechanical and morphological properties.

2. Materials and methodology

2.1. Materials and purification

Two well-known clay minerals samples were selected for this work: a grey blackish kaolinitic plastic material (referred as *Ba*) collected from Bamessing area (Nord-West Region, Cameroon), and a whitish sand-rich kaolinitic clay (referred as *Va*) from Mvan (Center Region, Cameroon). Wouatong et al. [22] have found that the main mineral phases present in *Ba* sample are kaolinite (~15–16%), quartz (~41–42%), illite (~15–22%), chlorite (~2–3%), with associated minerals such as goethite (~3.5–4%), gibbsite (~0.73–0.74%) and anatase (~1–1.3%). According to Djangang [21,26], kaolinite (~67%), quartz (~27%) and illite (~2.3%) constitute the main mineral phases of *Va* clay mineral, with anatase (~0.5%), ilmenite (~2.1%) and hematite as associated phases. The chemical composition of *Va* is ~67% of SiO_2, ~20% of Al_2O_3, and ~1.6% of Fe_2O_3. Before using raw *Ba* and *Va* clay minerals, they were soaked in water for two days and washed at 200 μm, decanted, dried on a plaster vase, then in an oven at 105 ± 2 °C, grinded and finally sieved at 63 μm. A sawmill by-product of Ayous (*triplochiton scleroxylon*) sawdust was collected in Yaounde (Cameroon) and used as porogent agent. The sawdust collected was washed, dried in the laboratory for 72 h then in oven at 150 °C for 24 h before grinding and softening ≤100 μm.

2.2. Characterization of raw materials

Plasticity parameters of *Ba* and *Va* raw clays (400 μm particle size) were determined using the Casagrande method, on a CNTROLS Model 22-TOO 30/G apparatus: the liquid limits were 99% for *Ba* and 35% for *Va* while the plastic indexes were 51 and 16 for the same materials respectively.

2.3. Preparation of ceramic candles

2.3.1. Investigation of the best formulation

To determine the optimum *Ba*/*Va*/*S* composite formulations with appropriate mechanical properties, parallepipedic (8×4×1 cm^3) shape bricks were elaborated. Ceramic matrixes were made by mixing *Ba* and *Va* clays in various weight ratios. They were referred as CM1 (100% *Va* +0% *Ba*), CM2 (75% *Va* +25% *Ba*), CM3 (50% *Va* +50% *Ba*), CM4 (25% *Va* +75% *Ba*) and CM5 (100% *Ba* +0% *Va*). CM3 and CM4 that exhibited good mechanical properties were reinforced with 0%, 5%, 10% and 15% of sawdust fiber. Each formulation was weighed and then mixed with around 10% of water, pressed on stainless mould with hydraulic press. The briquettes were first dried in open air at room temperature during three days, then in oven at 105 ± 2 °C for 24 h. Dried specimens were fired respectively at 800, 900 and 1000 °C in an electrical furnace (Insuni-Mic model) at a heating rate of 3 °C/min; during 270, 300 and 330 min with the holding time of 2 h for each temperature. The samples were free cooled in the furnace up to 25 °C.

2.3.2. Design of candles

Purified clay materials were weighed in appropriate ratio, mixed and then homogenized by adding water to control the plasticity. Mixed plastic paste was shaped on an electrical tower (Shimpo Whell 21 model) rotating at 210 tr/min. Considering the linear shrinkage of 4.3% maximum (obtained from the previous briquettes specimens), the dimensions of green cylindrical ceramic candle were as follows: height 10.5 cm, thickness 0.53 cm and diameter 4.16 cm. Green candles were kept on a plastic bag during 4 days to allow the progressive migration of water [23], dried on open air during two weeks, then fired in a furnace.

2.4. Characterization of parallelepipedic bricks and ceramic candles

The dimensions of parallepipedic dried bricks (L_d, l_d, e_d), fired bricks (L_f, l_f, e_f) and cylindrical candle ceramic filters were recorded with vernier calipers (10^{-1} precision). For each formulation of bricks, many samples were manufactured. The linear shrinkage after fired was obtained using the formula $100\times(L_d-L_f)\times L_d^{-1}$. The bulk density of elaborated ceramic (in g/cm^3) was obtained by calculating the ratio of the fired specimen weight (M_f) to its volume (V_f). Fired ceramics were weighed and soaked in pure water fo 24 h (M_w), and the apparent porosity of samples was determined according to Eq. (A.1) [20,24]. The flexural strength of specimens was recorded on a three points bend test IGM hydraulic press (minimum strength value 0.02 kN) and computed according to Eq. (A.2) [17]. All tests were carried out at a rise speed of 0.06 mm/min.

Ba and *Va* raw clay minerals samples and powders (80 μm particle size) of CM3 and CM4 fired at 900 °C were characterized using Fourier Transformed Infra-Red spectroscopy on a Bruker alpha-p spectrometer. Thus, about 2 mg of each sample were mixed with 200 mg of KBr to form pellets that were analyzed in absorbance mode.

Microstructures of ceramic filter were observed on a Scanning Electron Microscope Philips XL30.

The pore size distribution and the average pore diameter of sintered candle filters were determined using Mercury Intrusion Porosimeter (Autopore IV 9500 V1.09) working from 0.10 to 33000.00 psia, corresponding to pore size from 353 to 0.01 μm.

Hydraulic permeability of candles were evaluated by simply collecting the flow volume over time of non soaked filters, filters soaked during 1 d, and filters soaked during 5 days. For this procedure, a known volume of water was poured on the filter suspended on gallows, the filtered water flowing through the funnel was collected in a burette and the volume was recorded after each 10 min. Additional perme-