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#### Research paper

# Elaboration and characterization of materials obtained by pressing of vermiculite without binder addition

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#### ABSTRACT

Vermiculite materials were obtained by uniaxial pressing of potassic vermiculite powders obtained by sonication without any binder addition. The vermiculite powders, made of aggregates of particles with nanometric thickness and micrometric *in plane* dimension, were pressed in the range 17.7–80 MPa at room temperature and 200 °C, and further annealed in the range 400–800 °C. Pressing powders at 200 °C instead of 25 °C allowed the slight increase of the density of the formed materials (from 1.9 to 2 g·cm<sup>-3</sup>) due to the desorption of the water molecules from the interaggregate and interparticle spaces, allowing a higher densification. The density was also increased by tailoring the particle size distribution. The pressed materials were formed of oriented arrangement of vermiculite aggregates. The porous structure, characterized by mercury porosimetry, scanning electron microscopy, and small angle X-ray scattering, was modelled by oriented oblate spheroidal pores formed in the voids between the stacked aggregates organized in a structure possessing a cylindrical symmetry. The porous structure was found to vary with the pressure and the annealing temperature.

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#### 1. Introduction

Vermiculite, a lamellar hydrated aluminium iron magnesium silicate, has the property to exfoliate, when submitted to a thermal shock, due to the generation of steam in the interlayer spaces (Hindman, 1994; Huo et al., 2012; Marcos and Rodríguez, 2010; Valaškova and Simha Martynkova, 2012). In its exfoliated state, vermiculite possesses very interesting properties such as low bulk density, low thermal conductivity and comparatively high melting point (1240 °C–1430 °C), high specific surface area and good cation exchange properties (Potter, 2003). Its other benefits are oxidation resistance, thermal stability at high temperature and environmental safety (Hindman, 1994; Potter, 2003). For all these appealing features, exfoliated vermiculite remains one of most used clay mineral as evidenced by its number of applications reported in the literature (Harvey and Lagaly, 2013).

Powdered vermiculite was for example used as adsorbent of various pollutants in the field of water treatment (Kehal et al., 2010; Rytwo et al.,

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2009), as inorganic filler for the design of clay polymer nanocomposite (CPN) possessing improved mechanical properties (Du et al., 2003), or sealing properties (Takahashi et al., 2006), as lightweight porous filler for the production of heat-insulating refractory components (Valášková et al., 2009) or as material to produce thin inorganic films, coatings or foams (Minker-Villemin et al., 1998).

Among the different methods reported in the literature to obtain small sizes of vermiculite particles are ultrasound treatments. Several recent studies reported the use of low frequency ultrasounds to obtain micrometric and submicrometric particles from larger vermiculite samples (Ali et al., 2014; Nguyen et al., 2013; Pérez-Rodríguez et al., 2002; Poyato et al., 2012; Wiewióra et al., 2003). Those low size particles might be used as additives for the elaboration of various items. Many compact materials containing vermiculite powder were manufactured by different techniques to produce articles of various sizes and shapes. Some materials were produced by extrusion of mixtures containing different clays and vermiculite (Robinson, 1958). Fly ash based fibrereinforced phenolic nanocomposites filled with vermiculite were manufactured by compression under 15 MPa of a mixture of these different components heated at ~150 °C (Satapathy et al., 2011). Paper-like materials mainly composed of vermiculite were elaborated and cut at a desired shape to produce gaskets for sealing technology





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(Hoyes and Bond, 2007; Rautanen et al., 2009). The preparation of many other composite laminate materials containing vermiculite and an organic binder (Conner and Dumas, 1991) or an inorganic additive (Wiker and Work, 1992) was also patented. These materials were mainly prepared by laminating processes. A wide variety of forms of vermiculite products were also elaborated by moulding processes (McAloon et al., 1985). Vermiculite was always associated to at least one additive, either organic or inorganic in order to improve the properties and increase the performances of the different produced materials.

The objective was to develop a forming process in order to prepare compact vermiculite materials by uniaxial pressing in a mould, without any binder (nor organic neither inorganic). Thus, the roles of powder granulometry (millimetric and micrometric particles), temperature ( $T \leq 200$  °C) and pressure (P < 90 MPa) on the materials forming were investigated. The hot pressed materials (at 200 °C) were also annealed up to 1000 °C. The prepared materials were characterized in terms of texture, by scanning electron microscopy (SEM), small angle X-ray scattering (SAXS), mercury porosimetry, pole figure and density measurements. The hydration–dehydration of the materials was studied in order to better understand the influence of heat on the forming process.

#### 2. Experimental

#### 2.1. Materials

The starting vermiculite (purified exfoliated "Granutec® E", fine grade millimetric plates < 2 mm, originating from Yuli, China was purchased from CMMP, Paris, France). Vermiculites from the deposit were previously characterized by Huo et al. (2011, 2012). Particle size was reduced by milling to about 0.5 mm. The chemical composition of a half-unit cell of the raw vermiculite was  $(Si_3Al_1)(Mg_{2,62}Fe_{0,32}Ti_{0,06})$ O<sub>10</sub>(OH)<sub>2</sub>K<sub>0.45</sub>Ca<sub>0.08</sub>. Potassium chloride was used to saturate vermiculite in order to obtain a weakly hydrated initial material to improve the stability in further heat treatment. After potassium exchange, the average chemical composition of a half-unit cell calculated from elemental analysis was (Si<sub>3</sub>Al<sub>1</sub>)(Mg<sub>2.62</sub>Fe<sub>0.32</sub>Ti<sub>0.06</sub>)O<sub>10</sub>(OH)<sub>2</sub>K<sub>0.61</sub>. In order to produce micrometric and submicrometric particles (Ali et al., 2014; Nguyen et al., 2013), hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>, 35%) dispersions (55 mL) of a weighted amount of vermiculite (0.55 g or 3.85 g) were sonicated in a conical glass double-jacketed reactor cooled at room temperature, using a Sonotrode (20 kHz, 350 W, Ti ultrasonic probe, Sonics and Materials, amplitude 43 µm, acoustic power 56 W). The solid/liquid (S/L) ratio and the sonication time were varied in order to modify the particle size distribution. Powders were obtained by sonication of vermiculite dispersions prepared at two different S/L ratios (7% and 1%) during different sonication times (5 h and 12 h).

The sonication experiments of vermiculite dispersions were also conducted for 5 h in an ultrasonic tube (35 kHz, 400 W, Sonitube® from Synetude Chambéry France, active volume 78 mL, internal diameter 20 mm) supplied by a continuous closed flow (1 L/min) of 2 L of  $H_2O_2$  (17%) vermiculite dispersions (S/L = 7%). During this sonication, the temperature of the vermiculite dispersion was maintained at 35 °C using a double-jacket glass container (2.5 L) cooled by circulation of fresh water.

The particle size distributions of the dispersions were measured with a Mastersizer 2000 particle size analyser (Malvern Instruments, range  $0.02 \,\mu$ m to 2000  $\mu$ m).

#### 2.2. Elaboration of compressed vermiculite materials and annealing

After sonication, the dispersions were evaporated for 12 h at 80  $^{\circ}$ C and the dried vermiculite powders were used to elaborate materials. The vermiculite powder (about 500 mg) was pressed at room

temperature or at 200 °C under various pressures for 3 h in a hollow cylindrical mould to form hollow cylindrical pellets (thickness  $\approx 2$  mm, external diameter: 13 mm, hole diameter: 5 mm). All the materials were manufactured through the application of a uniaxial pressure using a MTS press (Insight 50). For hot pressing, the powder was first pressed at room temperature for 15 min and the pressure maintained, then the mould was heated by a ribbon heater (4 °C/min, for 45 min) and maintained for 2 h at 200 °C. After 2 h at 200 °C, the pressure was released and the sample turned out of the mould. Some of the formed materials (after the pressure was released) were heat-treated in the range [400 °C–1000 °C] for 10 h, in a muffle furnace (4 °C/min).

### 2.3. In situ study of dehydration and re-hydration (thickness measurement and dilatometry)

The dehydration of the vermiculite material was studied in situ during hot pressing at 200 °C and 80 MPa. For this experiment, the mould (described in Section 2.2) was filled with vermiculite powder (S/L = 7%, sonication time: 5 h) and pressure was maintained at 80 MPa using a MTS press (Insight 50), the mould was heated at 200 °C (<4 °C/min) using a ribbon heater and further cooled at room temperature. During this thermal cycle, the thickness of the sample was measured using a video displacement sensor (Apollor, precision:  $\pm 5 \mu$ m). The evolution of the radial dimension of a hot pressed (80 MPa and 200 °C) pellet (S/L = 1%, sonication time: 5 h) was also studied as a function of temperature by dilatometry (ODHT, Expert System Solutions) from room temperature to 1000 °C (heating rate: 5 °C/min).

#### 2.4. Characterization by X-ray powder diffraction

The powder from a raw pressed vermiculite material and further heat-treated in the range [600 °C–1000 °C] for 10 h was analysed by X-ray diffraction (XRD) for phase identification using a Bruker D8 Advance diffractometer ( $\lambda$ CuK $\alpha$  = 1.5418 Å) in a Bragg–Brentano ( $\theta$ ,  $\theta$ ) mode (from  $2\theta$  = 3° to 68°).

The heat treatment of a pellet pressed at room temperature at 200 °C and 400 °C was studied using a XRD Thermo Electron ARL'XTRA diffractometer ( $\lambda Cu_{K\alpha} = 1.5418$  Å, Si(Li) solid detector) in a Bragg-Brentano ( $\theta$ ,  $\theta$ ) mode. The evolution of the powdered vermiculite (30–60 mg) from pressed pellet (at 200 °C) was characterized in situ by XRD under controlled relative humidity on the same diffractometer. The humidity and the temperature were controlled by coupling a humidity generator (VTI HR 200) to a hygrometry chamber (Anton Paar HTK 1200R). XRD patterns were collected from  $2\theta = 2^{\circ}$  to 64° by step of 0.05°/10 s, after establishing an equilibrium period of 1 h prior to each measurement.

#### 2.5. Characterization by mercury porosimetry

Porosity and pore size distribution of the samples were measured with a mercury porosimeter (AutoPore III series 9400, Micromeritics Instrument Corporation). Before measurements, the samples were dried at 110 °C for 12 h and outgassed at  $10^{-3}$  mbar. Mercury intrusion tests were performed for pressures ranging from 0.005 MPa to 198 MPa (or pore diameter from  $6.3 \times 10^{-3}$  µm to 250 µm). The intrusion volumes were measured at stepwise increasing pressures, allowing equilibration at each pressure step. The presented data are averages from three replicated tests. The volume V [m<sup>3</sup>·kg<sup>-1</sup>] of mercury intruded at a given pressure P [Pa] gave the pore volume that can be accessed. Assuming cylindrical pores, the intrusion pressure was translated on equivalent pore diameter D [m], following the Washburn equation: D =  $-4\gamma \cos\theta/P$ , where  $\gamma$  is the mercury surface tension (0.485 N·m<sup>-1</sup>) and  $\theta$  is the mercury/solid contact angle (141.3°). The skeleton and the apparent densities of the pressed

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