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# Preparation of magnetic porous ceramsite and its application in biological aerated filters

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ABSTRACT

Ceramsite plays a significant role as a biological aerated filter (BAF) in the treatment of wastewater. In this study, a mixture of goethite, sawdust and palygorskite clay was thermally treated to form magnetic porous ceramsite (MPC). An optimization experiment was conducted to measure the compressive strength of the MPC. X-ray diffraction (XRD), scanning electron microscopy (SEM), and polarizing microscopy (PM) characterized the pore structure of the MPC. The results show that a combination of goethite, sawdust and palygorskite clay with a mass ratio of 10:2:5 is suitable for the formation of MPC. The compressive strength of MPC conforms to the Chinese national industrial standard (CJ/T 299-2008) for wastewater treatment. The SEM and PM results also show that the uniform and interconnected pores in MPC were well suited for microbial growth. The MPC produced in this study can serve as a biomedium for advanced wastewater treatment.



## 1. Introduction

Porous ceramsite is an effective media used in biological aerated filters (BAFs) for the treatment of wastewater [1]. The characteristics of porous ceramsite are related to the initial capital outlay, process design, and operation mode of BAFs. These characteristics also improve the efficiency and daily running costs such as backwashing and air influx. Mineral supports for porous ceramsite include zeolite, clay, sand, macadam, coke, anthracite coal, and plastic materials, such as polyethylene and polystyrene [2,3]. Studies in China during the late 1980s indicated that BAFs which contained expanded clay as a biofilm support could achieve superior substrate removal compared to sand and plastic media of similar dimensions [4].

Sawdust is a byproduct of timber processing and extraction. In China, the annual production of sawdust amounts to nearly 180 million tons and is increasing by 6–8% per year [5]. The generation of sawdust in earlier years has resulted in huge accumulations in many parts of China. The management of sawdust waste is a heavy economic burden to industry [6], and it is important to utilize or add value to waste sawdust. Lu et al. [7] reported that at high sintering temperatures the lignin and cellulose in sawdust can be converted into a carbonaceous porous material which could be used as a cellular material. However, to the best of our knowledge, only a small number of studies have reported the use of sawdust as a porous material [8].

Palygorskite is a natural magnesium-aluminum silicate clay mineral with a diameter of 30–50 nm. It is known to contain continuous two-dimensional tetrahedral sheets, but differs from other layered silicates due to its lack of continuous octahedral sheets. The tetrahedral basal oxygen atoms invert apical directions at regular intervals and thereby forming talc-like ribbons. It has a high surface area, viscosity, and porosity as well as significant thermal resistance and chemical inertness. In China, palygorskite resources are abundant in the eastern part of Anhui Province and western part of Jiangsu Province. It is an excellent absorbent, and recent attention has focused on the utilization of palygorskite [9–11].

To the best of our knowledge, there is no study investigating palygorskite for the production of ceramsite. The compositional and structural variations of ceramsite obtained in previous studies were attributed to many factors such as sintering temperature, sintering time, and the ratio of palygorskite/goethite. However, no

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studies have yet been conducted to investigate the impact of the specific and important constituents on the characteristics of ceramsite. As one of these components, palygorskite may strongly affect the bloating behavior and crystal formation of ceramsite during the heat treatment process.

The application of magnetic materials to solve environmental problems has received considerable attention in recent years due to their ability to separate substances in a liquid medium after adsorption [12–16]. Although magnetite ($Fe_3O_4$) has been used to precipitate heterogeneous calcium phosphate, its use in the context of environmental pollution has not received much attention. Scarce information is available on the potential application of magnetite as a phosphate sorbent [17]. Magnetized BAF has been used in treating wastewater, mostly to separate solids or attached microorganisms from effluent. Reports indicate that there was an increase in bacterial activity during the separation process [18].

The selection of a suitable BAF medium is critical in process design and operation if the required effluent standards are to be met. The characteristics of the filter media can significantly affect the initial investment and operational costs. The selection of the BAF media will depend on many factors, including its resistance to microbial degradation, its mechanical strength, the type of fluid used, its surface characteristics, and its cost. In this study, the MPC was prepared by sintering a mixture of goethite, sawdust, and palygorskite. Sawdust was treated at 400–800 °C to produce a desirable porous material. During sintering, goethite is transformed into magnetite in the presence of $H_2$ gas emitted during pyrolysis at 400 °C [19]. The formation of an interior porosity improves microbial growth. This study may lead to a promising new method for the preparation of biofilter carriers.

To validate this hypothesis, the present study has been conducted to: (i) utilize palygorskite/goethite and sawdust for the production of ceramsite; (ii) investigate the effect of palygorskite and sawdust on the physical characteristics (porosity) of ceramsite; (iii) characterize the ceramsite within the optimal content ranges of palygorskite/goethite and sawdust via thermal analysis, morphological structure analysis, X-ray diffraction, and compressive strength; (iv) analyze the sintering mechanisms; and (v) establish effective evaluation parameters.

## 2. Materials and methods

### 2.1. Material

The raw palygorskite came from Crown Hill located in the city of Mingguang, Anhui province, China. Sawdust was sampled from the city of Hefei, Anhui province, China. Goethite was obtained from the city of Zhenjiang, Jiangsu province, China. The particle size after extrusion, cutting, and crushing was less than 0.074 mm. Commercial ceramsite (CC) was obtained from the city of Ma'anshan, Anhui Province, China. The characteristics of MPC and CC are specified in Table 5. MPC were superior to CC in many ways, including porosity, specific surface area, and apparent density (Table 5).

### 2.2. Preparation of magnetic porous ceramsite (MPC)

The preparation of MPC includes the following steps:

(1) The mixture of goethite, sawdust, and palygorskite with the experimental mass ratio was placed in a small coating machine which produced round granules with a diameter of 7–13 mm.
(2) The products were dried at 110 °C.
(3) The composite iron oxide particles were put in a 2 L quartz tube reactor and heated at 500, 600, 700, and 800 °C. Each of these temperatures was maintained for 0.5–4 h under a $N_2$ atmosphere at a flow rate of 30 mL/min.
(4) The annealed MPC was cooled to room temperature over 12 h.
(5) The synthesized MPC was stored in a vacuum desiccator for future analysis.

### 2.3. The application of MPC in biological aerated filters (BAF)

Backwashing is a very important step in BAF operation. BAFs need to be optimized to reduce the water consumption and energy cost of backwashing. To achieve this, the current methods attempt to reduce the apparent density of biofilter carriers. The increase in apparent density facilitates the merger of well-developed microbial membranes with biofilter carriers in the BAF. This increases the inefficiency of BAF backwashing and may result in significant BAF short-circuited current. In addition, it will create to excessive BAF backwashing. These effects can lead to excessive biofilm detachment which can result in deterioration of effluent water quality.

In this research, magnetic porous ceramsite (MPC) is attracted to the electromagnetic iron-separator (Fig. 1). The experiments showed that the electromagnetic iron-separator can shift the MPC in the BAF. The MPC is moved to a certain height and then the electricity supply is switched off and the MPC sheds biofilm. At the same time, the MPC collide into each other and accumulate to form voids that facilitates regeneration ability. This method of making MPC voids obviates the need for backwashing, and the restoration process is much faster than combined action gas–water backwashing.

### 2.4. Characterization

The multi-point Brunauer–Emmett–Teller (BET) surface area of MPC was measured using a Quantachrome Nova 3000e automated surface area analyzer. X-ray fluorescence (XRF) chemical composition was measured on a Shimadzu XRF-1800 with Rh radiation. X-ray diffraction (XRD) was performed using a Rigaku powder diffractometer with Cu Kα radiation. The tube voltage was 40 kV and the current was 100 mA. The XRD diffraction patterns were taken in the range of 5–70 °C at a scan speed of $4° min^{-1}$. Phase identification (Search-Match) was carried out by comparison with those included in the Joint Committee of Powder Diffraction Standards (JCPDS) database. Elemental analyses of the sample were carried out by a VARIO ELIII analyzer (Elemental analysis system Co. Ltd., Germany). Magnetic susceptibility analyses of the samples were carried out via a Bartinton MS2 analyzer.

The thermal behavior of samples was examined by DTA-TGA7300 using an EXSTAR simultaneous DTA-TGA7300 analyzer while the samples were heated at a rate of 8 °C/min from 20 to 700 °C with nitrogen atmosphere at 100 mL/min. Samples ranged from 4 to 10 mg in mass, and they were compacted into a Pt–Rh crucible with 20 taps.

In the biological structures analysis, selected MPC were sputter coated with gold and its surface morphology was examined using a Scanning Electron Microscope (SEM, Philips XL30 ESEM). Biofilm determination was performed as previously described [20]. The growth of biofilm was determined according to the methods available in the literature [21]. The physical characteristics of the MPC samples were measured in accordance with the sandstone pore structure method of image analysis. Archimedes law (i.e., any object placed in a fluid displaces its weight and an immersed object displaces its volume) is an accurate method for porosity measurement that we utilized [22].