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Granulation processing parameters on the mechanical properties of diatomite-based porous granulates

Yujing Liu, Dagobert Scharf, Thomas Graule, Frank J. Clemens *

Laboratory of High Performance Ceramics, Swiss Federal Laboratories for Materials Science & Technology (Empa), Duebendorf 8600, Switzerland

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ABSTRACT

The development of cheap natural porous diatomite into catalyst supporting materials has obvious technical interests. High-intensity granulation process in the presence of clay-based inorganic binder system can be used to preserve the porous network of the diatomite structure and enhance mechanical stability of achieved granulates. In this work, via tuning the granulation parameters, such as pan rotation speed, rotation modes between the pan and the impeller, as well as the factor of granulation time, the size distribution and the mechanical stability of formed granulates were adjusted. Specifically, the pan rotation speed can assist the change of granulate growth mechanism and thus affect their mechanical attrition resistant ability; the rotation mode between impeller and pan has an influence on the powder distribution inside of the pan and the modification of granulate growth rate can be observed; and a certain prolongation of granulation time has no influence on granulates mechanical stability but only increases the granulate size.

1. Introduction

Diatomite is categorized as a type of natural porous materials. It consists principally of the fossilized skeletal remains of diatoms, with $SiO_2.nH_2O$ and crystallized silica as the main composition. Thanks to the large varieties of the species of diatoms and their complex organizations, diatomite possesses a wide range of pore size distribution, from nanometer to micrometer scales [1,2]. The porous structural characteristic and bio-compatible chemical composition, together with obvious advantages of low price and rich abundance, provide diatomite with wide applications in catalysts, separation filters, bio-technical device scaffolds and other related areas[3]. However, for diatomite application as catalyst supporting materials using in fluidized bed reactors (e.g., chemical looping combustion process, methanation industry, et.al), a requirement on its mechanical stability is also of great necessity in addition to the sound needs on its porous network.

Efforts to enhance the mechanical stability of porous diatomite-based materials are mainly lying on the tuning of sintering process at elevated temperatures. Aderdour et al., [4], Zhang et al., [5], Yilmaz and Ediz [6] and Degirmenci and Yilmaz[7] have reported the effects of sintering temperature on mechanical strength improvement and microstructure variation of diatomite or diatomite-based composites. Another approach to enhance the mechanical property of diatomite-based macroscopic structures is via the addition of so called binding materials. Starch [8] and sodium carbonate [9] were added to 'glue' diatomite together before and after the sintering process. Beside to the previous work, Van Garderen [10] observed that both the sintering temperature and clay content (as binding material) affect the attrition resistance and porous properties of extrusion fabricated diatomite granulates. However, the properties of granulates are not only decided by their chemical compositions and microstructure but also strongly influenced by the processing conditions.

Wet granulation is defined as a process to form granulates during the addition of a liquid or binding solution into a continually agitated powder system [11,12]. It is widely applied and plays important roles in the processing of pharmaceutical, agricultural products, minerals and specialty chemicals [13,14]. In many cases, this process is carried out by high-intensity granulators, which incorporates a container, an impeller in the base and a vertically mounted chopper. The impeller provides the mechanical energy required for mixing the solid contents, and the chopper is designed to break up granules that are formed. Processing conditions [11–20] of high-intensity granulators, such as operation speeds of the impeller and/or container, chopper speed, granulation time, etc., play critical roles on the characteristics (the published work mainly focused on granulate size distribution) of fabricated granulates. Thus, in order to produce granulates with targeted properties, one has to carefully control the processing conditions of the high-intensity granulator.

In this work, an optimized extrusion recipe based on the previous work of van Garderen [10] was selected to perform our investigation of the influence of the granulation parameters on the properties of

* Corresponding author at: Empa, Swiss Federal Laboratories for Materials Science and Technology, Überlandstrasse 129, 8600 Dübendorf, Switzerland. Tel.: +41 58 765 4821; fax: +41 58 765 6950.
E-mail addresses: yujing.liu85@gmail.com (Y. Liu), frank.clemens@empa.ch (F.J. Clemens).

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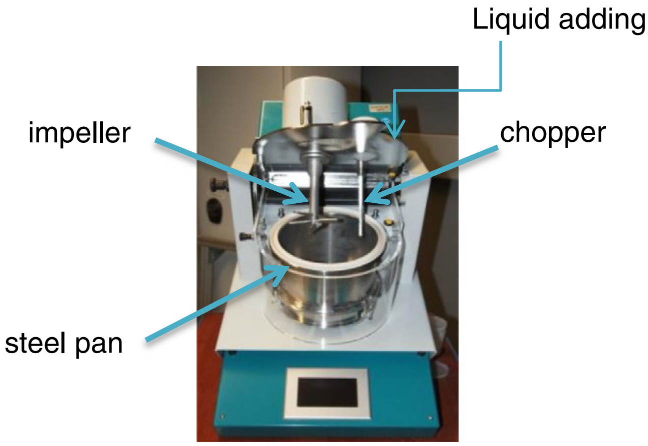

Fig. 1. Eirich high-intensity granulator (impeller diameter 7.7 cm, steel pan diameter 16.5 cm).

porous diatomite granulate. Aspects to evaluate fabricated granulates include the size distribution, mechanical stability (characterized with attrition resistance) as well as the porous characteristics.

2. Materials and processing method

2.1. Materials

Diatomite-based porous granulates are prepared from a mixture of two different types of diatom cellites, namely, standard cellite and filter cellite (CELITE, World Minerals Inc., USA). These two diatom cellites share the similar silica-based chemical composition (91.1% SiO_2 for standard cellite, 89.0% SiO_2 for filter cellite) but slightly differ each other on the median pore sizes and the morphology [21]. According to the recipe of van Garderen [10], a certain amount (28 wt.%) of clay kaolin EKA-S (less than 180 μm, Amberger Kaolinwerke, Germany), used as a plasticizer and as an inorganic binder to achieve sufficient mechanical properties, and diatom cellites (34 wt.% of standard cellite and 38 wt.% of filter cellite) were dry mixed. Additionally, organic oil contained liquid phase (78 wt.% of solids) was added to start the wet granulation process.

2.2. Granulation process

The diatomite-based natural porous granulates are fabricated in a 1 L Eirich high-intensity granulator (Maschinenfabrik Gustav Eirich GmbH & Co KG). Steel pan (container), impeller and chopper are important components of this machine, as shown in Fig. 1. During the granulation process, the solid contents (174.7 g powders of two different diatomites and clay kaolin) were first thoroughly mixed in the steel pan, and then the liquid phase was added in the form of sprays via a pressure controlled spray generator (pressure 3 bar). The time to add liquid phase is 2 min. Within this period of time, secondary units of nuclei come into birth, and we thus name it as the nucleation time. Afterwards, granulate starts to grow, and a period of 6 min was set as the granulating time. To investigate the effect of granulation time on the properties of generated granulates, we varied the granulation time from 3 min to 9 min.

To study the effect of pan rotation speeds (85 rpm and 170 rpm), the rotation speed of impeller has been kept constant: 1984 rpm for the period of nucleation formation and 6200 rpm for the period of granulate growth. The steel pan (positioned horizontally) can only rotate in a clockwise direction, while the impeller inside can rotate both at clockwise and anti-clockwise directions. Thus, two different rotation modes can be generated when we combine the rotation directions of the steel pan and impeller in different ways: counter current rotation mode, where both the pan and the impeller rotate clockwise; cross current rotation mode, where the pan rotate clockwise but the impeller rotate anti clockwise. It is worthwhile to mention that the counter current rotation

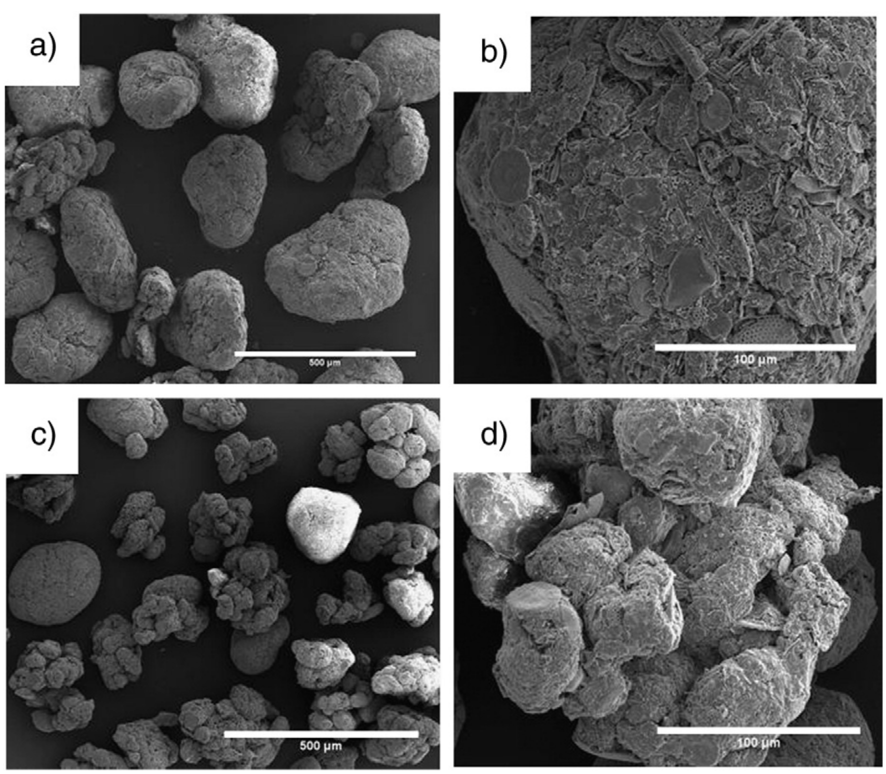

Fig. 2. Top view SEM images of sintered granulates fabricated under different pan rotation speeds: (a and b) granulates fabricated at pan rotation speed 85 rpm, (c and d) granulates fabricated at pan rotation speed 170 rpm. The scale bars in panels a and c are 500 μm, in b and d are 100 μm.