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Attrition of methanol to olefins catalyst in jet cup

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ABSTRACT

Attrition of catalyst in a fluidized bed reactor is an inevitable issue especially in a commercial unit. Methanol to olefins (MTO) is becoming one of the main stream technologies for light olefins production. The attrition of MTO catalyst, however, received little attention. This study is focused on the attrition behavior of MTO catalyst in jet cup at high temperature. The influence of test time, inlet gas velocity, and temperature on MTO catalyst attrition was studied. It is found that the Gwyn formulation can well represent the relation between attrition index and test time. Our results show that jet cup can retrieve results quantitatively comparable to high velocity gas jets method while significantly shortening test time. It is also found that the inlet gas velocity has considerable influence on the MTO catalyst attrition, and the relation between inlet gas velocities and attrition index can be described by a power index of 3.7. Similar to high velocity gas jets experiments the attrition index manifests a maximum with the increase of temperature. But the temperature corresponding to the maximum attrition index shifts from 300 °C in high velocity jets tests to 100 °C in jet cup experiments. An analysis based on SEM pictures indicates that the transition of attrition mechanism is responsible for this shift. An empirical correlation has been presented for MTO catalyst attrition in jet cup, which shows good agreement with experimental data for inlet gas velocity from 88 to 158 m/s, and temperature from 100 to 500 °C.

1. Introduction

Fluidized bed is one of the widely used reactors in chemical industry due to its excellent performance such as perfect fluid-solid contact, efficient heat and mass transfer between fluid and solid, uniform temperature distribution and good thermal stability in the reactors and so on. However, particles attrition and the wear of inner walls of the equipment are the highly concerned disadvantages especially for the pilot and commercial plants. Continuous movement of particles inside fluidized bed will cause frequent particle-particle and particle-wall collisions, which, in many applications, can result in severe decrease in particles' strength and eventually lead to surface abrasion and/or breakage of particles, wear of the equipment as well. Therefore, research on catalyst attrition in fluidized beds is of practical significance in catalytic process development. Methanol to olefins (MTO) is a recently developed catalytic process for on-purpose ethylene and propylene production with methanol as feedstock. MTO starting from coal was first commercialized in 2010 in China by Dalian Institute of Chemical Physics, Chinese Academy of Sciences. MTO is becoming one of the main stream technologies for light olefins production. In MTO process, fluidized beds were used as reactor and regenerator, sharing similarity with the fluid catalytic cracking (FCC) process in refineries. In the past decades, the attrition of FCC catalyst was a subject of intensive study [1–11]. The attrition of MTO catalyst, however, received little attention.

Typically catalyst attrition in fluidized bed reactors can be due to complicated mechanical, thermal, and chemical stress experienced by catalyst particles. Thus both material properties and operation conditions will influence the attrition rate of catalyst in fluidized beds. One of the challenges in catalyst attrition study is that the attrition process of catalyst in an industrial fluidized bed reactor can span months or even years. In laboratory the research with such a time scale, if not impossible, is unrealistic. In this regards, a variety of research methods [12] have been developed to study catalyst attrition in laboratory. For example, single particle impact experiments [13], shear tests [14], drum tests [15], high velocity gas jets [10,11,16] and jet cup [9,17,18]. Amongst these methods, high velocity gas jets and jet cup are commonly-used for bulk attrition testing of fluidized bed catalyst [9,19]. But there are certain differences between these two methods. In high velocity gas jets method, high speed gas flow passes through one or three holes of micrometer size in a distribution plate, agitating severe collisions between particles. In the jet cup method, gas enters the vessel tangentially. In this case particles will be dragged and move upwards spirally, experiencing frequent collisions with the inner wall of the jet cup. Weeks and Dumbill [9] and Zhao et al. [19] compared these two methods, and found that these two test methods are comparable in terms of attrition propensity ranking. But jet cup method requires

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fewer samples and can speed catalyst attrition-resistance determination. The early type of jet cup was cylindrical with small diameter, which had been improved afterwards by Cocco et al. [18]. Jet cup method has been used effectively to assess the attrition resistance of FCC catalyst [1,2,9], slurry bubble column reactor catalysts [20], spray-dried iron Fischer-Tropsch catalysts [21–24], oxygen carrier particles for chemical looping combustion [25,26] and so on.

In a previous study [27,28] we studied the attrition of commercial MTO catalyst by use of a high velocity air jet unit. It has been discovered that the attrition mechanism of MTO catalyst at room temperature is different from that at high temperature. Thus catalyst attrition resistance evaluation at room temperature might not reflect the situation at high temperature. In addition to that, it is suggested that at least 24 h is required for an attrition test in high velocity gas jets facility to achieve an equilibrium state for MTO catalyst. The purpose of the current study is to investigate the attrition of MTO catalyst in a jet cup at high temperature. Although the jet cup method has been used extensively, most of work was carried out at room temperature. We will focus on the comparison of our jet cup results with those obtained via high velocity gas jets, and try to establish an empirical correlation of MTO catalyst attrition index with jet cup operation parameters such as inlet gas velocity, temperature and test time.

2. Experimental

2.1. Jet cup attrition apparatus

Fig. 1 shows the jet cup experimental unit used in this work. Cocco et al. [18] studied particle motion in five different types of jet cups by use of cold flow experiments and CFD simulations. They found that the conical jet cup is more suitable for quantitative test of particle attrition, and testing results could be readily related to the attrition loss rates in fluidized bed cyclones. Therefore a conical jet cup made of stainless steel was adopted in our work. The jet cup unit can be heated up to 600 °C by a furnace. The top part of the settling chamber that was not enclosed by the furnace was covered with thermal isolation cotton. A preheater was installed to heat the compressed air the required temperature before entering the jet cup, which minimizes the effect of cool gas on catalyst attrition.

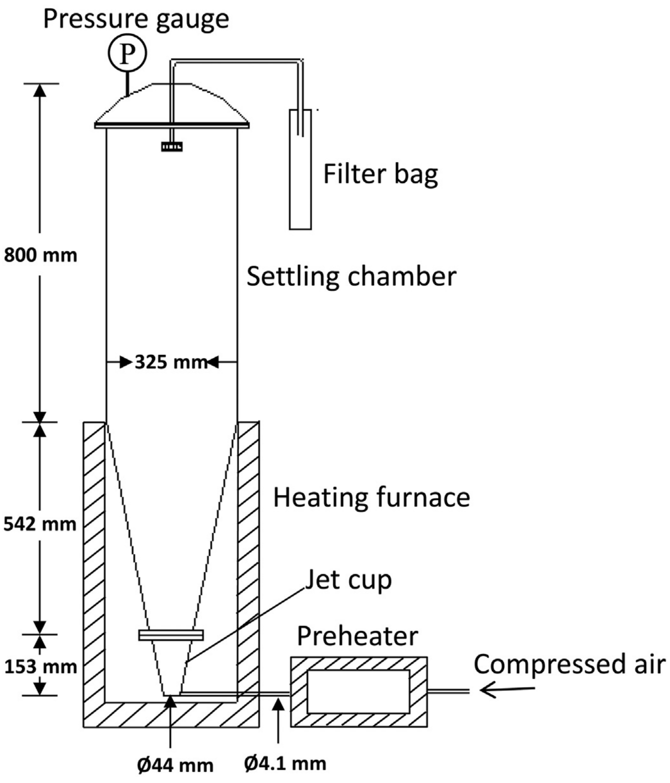

Fig. 1. Experimental apparatus.

The conical jet cup is 153 mm high, with diameter of 44 mm at bottom and 73 mm at top. A small tube of diameter of 4.1 mm is tangentially connected to the bottom of jet cup, serving as the gas inlet. The jet cup is attached to the settling chamber by flange connection. The settling chamber is cylindrical at the top and conical at the bottom. The diameter and height of the cylindrical part are 325 and 800 mm, respectively. The conical part is 542 mm high, with a small diameter of 86 mm at the inlet and a large diameter of 325 mm at the outlet.

2.2. Catalyst materials

Fresh commercial MTO catalyst was used in this work. Prior to each test, the catalyst sample was sieved to remove fines. The particle size distribution (PSD) of the sample is shown in Fig. 2. The loose bulk density is 0.75 g/cm^3, the median particle diameter (d_{p50}) and Sauter mean diameter (d_{p32}) areabout112 μm and 107 μm respectively.

2.3. Attrition measurements

The sieved catalyst sample was heated in a muffle furnace for 3 h at 600 °C and cooled down to room temperature in a vacuum desiccator. Then 100 g powder was weighed and charged into the jet cup. After the flange connection between the jet cup and settling chamber had been tightened, the gas flow started. Before the gas flow meter was adjusted to the desired value, a gas leakage check was carried out. Each test lasted for 3 h. There are three streams of catalyst particles that need to be collected after each test. The first steam is fine particles entrained from the outlet of settling chamber, which were collected by filter bags. The second and third stream are catalyst remaining in the jet cup and fines adhering to the inner wall of the unit. After each test, all three streams of catalyst were weighted and analyzed. A material balance analysis showed that the maximum fine loss was approximately 1.5% of the initial sample for all tests.

PSD of catalyst was analyzed by Malvern laser particle size analyzer (Mastersizer 3000). A total PSD of the sample after each test was calculated based on PSD of each stream of catalyst particles according to the weight fraction. The details were described in our previous publication [27]. The morphology of catalyst was observed by a scanning electron microscope (SEM, Hitachi™ 3000). In this work, attrition index (*AI*) is used to characterize the attrition of MTO catalyst. *AI* is defined as the weight percentage of particles smaller than 20 or 44 μm (expressed as AI_{20} and AI_{44}, respectively) in a sample after a certain duration of test.

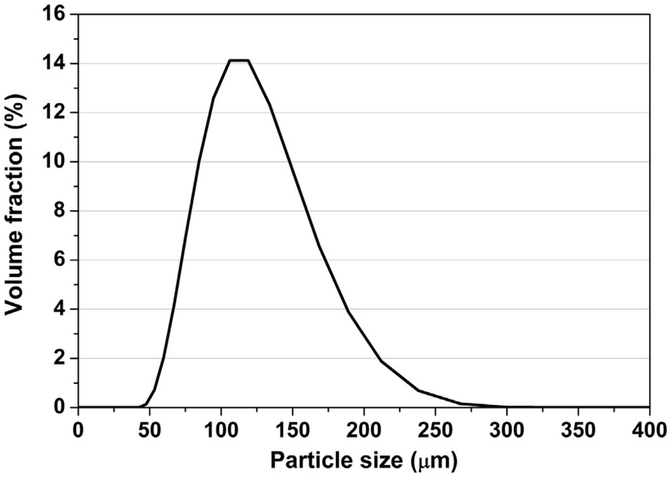

Fig. 2. Particle size distribution of the fresh catalyst sample.

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