Catalyst Classification According to Activity By Air Acceleration

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Abstract. Catalysts gradually get deactivated in the reactor due to deposition of carbon and metals on it which results in its density increasing. The moving catalysts with different activities have different acceleration in the pulsing air under the actions of various forces, which inspire us to develop an air acceleration classifier. According to the relationship between activity and density of catalyst found in this paper, theory of air accelerating classification was built and the particle dynamical equation was developed. Based on the theory of air acceleration classification, calculation was made with the Fluent software and EDEM software, based on the standard k-ε turbulence model and the Eulerian model. The coupled CFD-EDM simulation of material separation was performed to analyze the state of particle and column and obtain the statistics of particle in the classification column, which can help to study further of classification mechanism and particle movement in column and the experiment results are consistent with simulation. The maximum separating efficiency was up to 89%, which verified the possibility of air accelerating classification applied in separating different activity catalysts. The paper also studied the influence of particles’ shape and size for classification so that it can guide both experiments and engineering applications.

1. Introduction
Catalytic technology is the core technology of the chemical industry and more than 80% of the chemical industry is involved in catalytic technology [1]. During operation, catalysts are gradually get deactivated due to during an operation given the deposition of carbon and metals on the catalyst, so some of them. Numerous catalysts become hazardous waste in which a large part of them is discharged refining catalysts [2]. Carbon and metals deposited in the holes of the catalyst not only lead to catalysts’ deactivation, but also make their density increased, which results in a phenomenon that the density of lowly activity catalyst is lower than highly active catalyst for same catalyst [3]. The discharged catalysts are a mixture of catalysts with various activities. It wastes resources and leads to potential environmental hazards and causes difficulties in waste treatment.

For comprehensive disposal, the first step is separating discharged catalysts according to their activities. Wet separation methods modify the properties of waste catalyst due to the presence of water. Air separation is the process of material separation and transportation according to the differences of sedimentation motion after various forces acting on it. Traditional air separation is widely used [4, 5] and typically classifies particles in a rising or horizontal airflow. Many studies on air separation have
successfully achieved industrialization [6, 7]. The traditional air separation strengthens separation through a terminal velocity. However, this process has shortcomings, such as low accuracy, limited range of selection, and difficult to control. Pulsing air separation theory was proposed in the 1980s and used pulsing air to periodically accelerate particles which leads to accumulation of particle displacement difference, so that particles can obtain density-dominant separation. This method is mostly used to separate solid waste [8], vermiculite [8], fine coal [9], and a mixture of precious metals and concentrated catalysts after reforming and sintering tailings [10].

In view of the discharged catalysts characteristics of small size and different activity for different density, the separation technology based on activity difference was presented. Air acceleration classification is a technology using a pulsing air generator to produce a periodically pulsing air with specific waveform and large amplitude in the vertical separation column, and the particle in column will be accelerated periodically. Because the different activity particles with different densities, they will obtain different acceleration behaviors so that can be separated effectively. It can not only reduce the catalyst loss, but also reduce environmental impact, and serves as a valuable reference for the separation enhancement of particles.

2. Numerical Simulation

2.1. Model and simulation conditions

In order to study the process of particle classification in the column better, the CFD software FLUENT and EDEM were used to simulate the actual separation state through CFD-DEM coupling. The geometric model established by the ICEM software is same as the actual column. The total height of the classification column is 1950 mm and the diameter is 100 mm.

Based on multiphase flow and interactions between phases, the Eulerian model was selected as the basic model for numerical simulation. After calculation and analysis, the particles were in a turbulent state. When the shear stress changes, and the force exerted by the particles on the air flow will have a great influence on the turbulent motion of the air flow [18]. Therefore, the turbulence model is chosen to be a standard k-ε model with reasonable accuracy. And the drag model was set by writing and introducing a user defined function according to the law of air velocity generated by the pulsing valve and its drag coefficient.

The simulated materials are particles I and III. Particle I is fresh, spherical, highly active fluidized bed catalyst with activity of 100% and density of 1.612 g/cm³ while particle III is dedicated catalyst with activity of 50% and density of 1.909 g/cm³. Both particle size is 500 μm. Set the basic parameters of the simulation as feeding quantity 10 kg/h, feed ratio 1:1, maximum turbulent airflow speed 3.2 m/s, pulsation frequency 1.5 Hz, and air density 1.29 kg/m³. Particles generated by EDEM were added into the flow field simulated by FLUENT, and they were coupled to simulate the process in air flow acceleration classification column.

2.2. Simulation results

Due to the combination of gravity, drag force, and additional mass force, the particles' motion is complex. Take 0.5s as the starting time to extract images at intervals of 1 s. Figure 1 shows the movement and separation of simulated particles in an accelerated pulsating airflow field. The blue particles are highly active particles while the red particles are lowly active particles. At t = 0.5 s, the particles entering the column moved downwards under the action of gravity; at t = 1.5 s, particles began to appear the area above the inlet of the column; the particles oscillate under the action of pulsing air in column over time, the density of the highly active particles is smaller, so the force generated by the air is bigger than the gravity, and the particles show an upward movement tendency; the lowly active particles have a relatively high density, and the force generated by airflow was smaller than gravity, the particles show a downward trend as a whole. At t = 5.5 s, particles escape from the top and bottom outlets respectively, and blue particles are concentrated in the upper part of
the column. Due to continuous feeding, the color difference is not prominent. Therefore, the final highly active particles enter the upper hopper, and the lowly active particles enter the bottom hopper.

Figure 1. Motion and separation of particles at different time

Figure 2 records the change of particle velocity with time. Six particles were selected randomly to study particle velocity. Six particles added at different times, and periodic motions were performed with the pulsing air flow. When the airflow raised, the acceleration of particles increased from negative to positive and the velocity also gradually increased. When the airflow decreased, the acceleration of particles decreased and changed from positive to negative. The maximum velocity achieved by the lowly active particles was always less than the highly active particles while the minimum velocity of the lowly active particles was always more than the highly active particles. So the tendency of the highly active particles separated from the mixture and rising with the air become apparent, and the lowly active particles still fell down to the bottom hopper. The highly active particles are easier to be affected by the air than lowly active particles. In addition, there is a slight difference in the motion of the same kind of particles because the collision between the particles and particles and wall. However, it still conforms to the movement law of the particles in the flow field.

Figure 2. Velocity-time picture of highly/lowly active particles.

Since the feed port was located at $y=1400$, the particle added to column form the position near $y=1400$. The displacement of the particles in the Y-axis direction with time increasing is shown in Fig.
3. The time for the six particles to enter the device is different, but the overall law is the same. The displacement of particles showed a periodic change. The highly activity particles began to rise after 2-3 cycles falling, and raise to the feed port after 2-4 cycles. The cumulative displacement of highly activity particles was in the positive direction and they moved upwards to the top hopper. Meanwhile, the cumulative displacement of the low-activity particles was negative and they dropped down to bottom hopper. After about 9 seconds, the mixture achieved effective separation. For the highly active particles I₁, I₂, and I₃, there were differences in the displacement changes, but the overall movement tendency was consistent; the low-activity particles III₁, III₂, and III₃ were the same. The individual differences were due to the randomness and uncertainty of motion caused by collisions between particles and particles and walls.

![Figure 3](image.png)

**Figure 3.** Displacement-time picture of highly/lowly active particles.

3. **Experiment**

3.1. *Equipment and material*

The air acceleration classification device consists of a feeding device, an air supply system, a classification device and an exhaust gas process device. The scale of the laboratory apparatus is as follows: the total height of the sorting column is 1950 mm, diameter is 100 mm, screw feeder is 0 - 20 kg/h, pulsation frequency is 0 - 1.5 Hz, and air intake is 0-200 m³/h. In the laboratory, there are many factors affecting the actual material sorting. The feed rate was 8 kg/h, the pulsation frequency was 1.5 Hz, and the air flow rate was adjusted from 2.8 to 3.7 m/s.

Because it is difficult to obtain catalyst in uniform activity, the particles in various density were used to simulate different active catalysts according to the relationship between catalyst activity and density. The material was shown as Table 1. Different particles were mixed in half.

| Particle | I   | II  | III | IV  | V   |
|----------|-----|-----|-----|-----|-----|
| Density(g/cm³) | 1.612 | 1.788 | 1.886 | 2.117 | 2.262 |
| Activity | 100% | 75%  | 50%  | 25%  | 0%  |
| Size(μm)  | 500  |      |      |      |      |

3.2. **Results**

Taking both top hopper product and bottom hopper product into consideration, the highly active catalyst recovery ratio $E_{high}$ and the lowly active catalyst recovery ratio $E_{low}$ are needed to analyze comprehensively. Besides, the overall efficiency $E$ was also an important parameter to elevate the classification results. When the air velocity changes, the mass ration of production from top and bottom outlet will also change, which will have influence on results. Take particle I and III as an example and the results are shown as Figure 4. The highly active catalyst recovery ratio $E_{high}$ in top
production tend to decrease with air velocity rising. Conversely, the lowly active catalyst recovery ratio $E_{low}$ tend to increase. And the whole separating efficiency $E$ increased first and declined then and reach the optimal state at 3.36m/s where the $E_{high}$ and $E_{low}$ are both at high level. When the airflow velocity was in the range of 3.0-3.4 m/s, the total classification efficiency was above 70%, which proved that the material could still be effectively classified even in the case of air velocity fluctuations.

![Figure 4. Efficiency of mixed catalyst I and III](image)

Four different groups of materials were subjected to experiments. The classification efficiency is shown in Figure 5. The experimental results showed that the efficiency of airflow acceleration classification for all four groups was generally in a parabola form: increased first and then decreased. As the airflow velocity increased, the classification efficiency increases continuously; the maximum value was reached near the optimal airflow velocity; thereafter, with the air velocity continued to increase, the efficiency began to decline. Compared to particles in small activity difference, the particles in large activity difference obtain lager difference in acceleration kinetic energy which make them movement path more different and have less impact on experiment results when air flow is instable. Moreover, the operation region become larger to particles in large activity difference, which means stable separation efficiency and easy to separate. The best separation efficiency among these works is 89 % when the airflow velocity is at 3.4 m/s, the pulsing frequency is 1.5 Hz, and the feed rate is 8 kg/h.

![Figure 5. Efficiency of mixed catalyst at different activity.](image)
4. Conclusion

(1) Numerical simulations were performed on the process of air flow acceleration classification for 100% and 50% active fluidized-bed catalyst particles. The sorting process and the particle motion state obtained by CFD-DEM coupled simulation are obtained. The results show that the velocity of particles in the column are periodically changed. The overall motion of highly active catalyst first decreases and then rises. The overall activity of the low-activity catalyst tends to decrease.

(2) The efficiency of air acceleration classification generally increased first and then declined with the increase in air velocity and reached peak at the optimal air velocity. With the increase of activity difference between catalysts to classification, the efficiency is continuously increasing. The highest efficiency is up to 89.0% for particle I and V, and the efficiency for the simulation selected particle I and III is 80.1%. The experimental results are basically consistent with the numerical simulation results and it has been confirmed that the airflow acceleration classification can achieve the purpose of classifying catalysts according to their activities.

(3) The actual efficiency of airflow acceleration classification is affected by multiple factors. To achieve the industrial application, further experimental research is needed to explore the control parameters and improve devices.

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