

FLUIDIZED-BED JET MILL FOR CONTROLLING GRANULE MORPHOLOGY OF ULTRA-FINE SiC

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Abstract: Granule morphology exerts great influences on many properties of particle clusters. To obtain diversified particle shapes under different experimental conditions, theoretical analysis and experimental study were carried out on the processing of ultra-fine SiC (Silicon Carbide is a kind of semiconductor material) with fluidized bed jet mill according to the application situation. Special attention was paid to the main factors for controlling particle shapes. The shaping technique after pulverization process and the pretreatment techniques before ultra-fine pulverization process were also presented. The results show that well spherical or rhombus-like particles could also be obtained under varied experimental conditions of fluidized-bed jet mill. Experimental investigations into primary factors showed that particle shapes are correlated with jet mill structure (nozzle quantity), manipulated variables (comminuting intensity, gas-solid concentration, pressure, grinding time), and particle properties (granular size).

1. INTRODUCTION

Preparation technique of ultra-fine materials is necessary for the production of advanced applications. Solid materials, in the form of powder as the industrial raw materials, are required by the modern industry. The solid powder should be narrow in size distribution and low in impurity, and diversified granule morphologies are required based on the application environment. Granule morphology affects the particle cluster performance significantly in such factors as specific surface area, fluidity, filling, shape separation, chemical activity, covering ability of painting, liquid resistance of powder layer as well as fluidity resistance of particles to liquid. In engineering, different granule morphologies are required according to their applications. For example, a grinding powder should be spherical in shape and equal in volume; rhombohedra particles are needed because different shapes will lead to different

performances and breakage resistances; cement particles should be more spherical particles in order to improve the fluidity of concrete and lower the water consumption; large length-diameter-ratio is required for the aedelforsite powder while large diameter-thickness-ratio is demanded for the mica micro-powder. The controlling technique of granule morphology for the preparation of ultra-fine powder is a key subject faced in powder techniques. Ultra-fine materials grinding plant mainly consist of a jet mill and a classifier which has been applied in many fields, such as abrasive, ceramics, electronic materials, pharmaceuticals, etc. [1-4].

Jet mill is usually used in China, and it can ensure the best product quality of various dry ultra-fine powder particles with high-purity. The jet mill is characterized by simple technique for providing fine particle size, narrow granularity distribution, high-purity, total negative pressure system and no

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pollution. Therefore, the jet mill for processing ultra-fine powder particles has been developing quickly recently [5].

In China, systematic researches for controlling granule morphology are rarely reported in literatures nowadays. Both at home and abroad, the jets in fluidized beds can cause intense interparticle collisions, and this has been used successfully for milling of materials that are difficult to break or for producing very fine products [6-8]. Moreover, the construction of solid particle (material removed desirably) can be achieved according to the application [9]. However, the above-mentioned method for controlling granule morphology is rarely reported in China, which thus restricts its application. In order to obtain diversified granule morphologies according to specific particle properties and application environment, the paper aims to introduce the methods of fluidized-bed jet mill to control the granule morphology under different operation conditions in preparing ultra-fine particles and the shaping techniques for controlling granule morphology, as well as pretreatment techniques.

2. Fluidized-bed jet mills for controlling granule morphology

The structure of mill chamber is shown in Fig. 1. Solid feed is blown into the mill chamber by an air-pusher. High pressure air enters the mill through a number of nozzles placed around the cylindrical chamber. Then the collisions between the particles in the highly turbulent jets result in comminution in the mill chamber.

2.1. Grinding mechanisms of jet mill

In the process of jet milling, the particles are forced to impact with each other by air flow. Then the dispersed powder flow leaving the mill is split into coarse fraction which is recycle and fines which are collected in a filter as qualified product. The grinding of materials can be divided into brittle grinding and fatigue grinding according to analysis of asymptotic theory. Air flow grinding mainly consists of brittle grinding and auxiliary fatigue grinding. Large particles are accelerated to the required velocity through instantaneously intensive supersonic air flow and the collisions between particles occur at high velocity. So the produced stress value generated exceeds the brittle strength limit, resulting in brittle grinding for materials. The higher the velocity is, the larger the kinetic energy of particle is, and the effect of brittle grinding is notable. Fine particles are difficult to comminute because they are too fine to energize. The increased specific surface area leads to

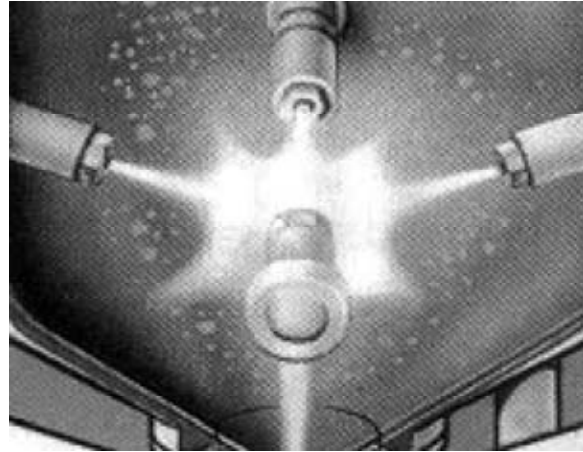


Fig. 1. Schematic diagram of mill chamber.

decreased cracks and defects. So the fine particles are only accelerated to shock, collide and frictionate with each other, producing notable plastic deformation. Cracks and avulsions occur in the inside of the materials, which leads to fatigue grinding when internal stress value reaches or exceeds the fatigue strength limit of particles. To assume that hard and brittle particles are absolutely flexible, the required velocity is named W when the particles collide freely under the shock condition.

$$W = \sigma \sqrt{\frac{1}{E\rho(1-\varepsilon^2)}} \quad (1)$$

Where σ is the strength limit of material and denoted by MPa; E is elastic modulus of material denoted by Pa; ρ is material density denoted by kg/m³; ε is the resume coefficient of particle velocity under the impact condition.

The parameter W is equal to W_1 plus W_2 when the particles collide oppositely. The parameter W is equal to W_1 subtract W_2 when the particles catch up with each other and then collide. According to formula (1), the required velocities to break materials are related to the mechanical performance of ultimate strength and elastic modulus. The greater velocity the particles get in the grinding chamber, the higher the grinding intensity is, the less collision frequency of a particle is, and the poor granule morphology is, and vice versa. The mechanisms of jet mill are not only collision grinding, but also friction. Friction forces produced by the relative motion of particles can remove superficial angularities of particles, so that more spherical particles can be obtained.

2.2. Preparation of silicon carbide

Granule morphology is closely correlated with the material properties after ultra-fine grinding. Diversified ultra-fine powder granularities and granule

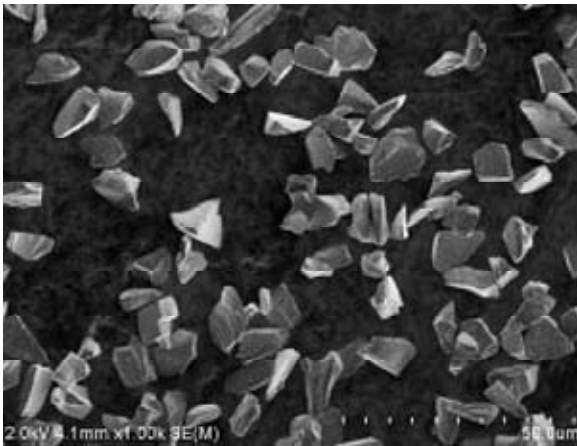
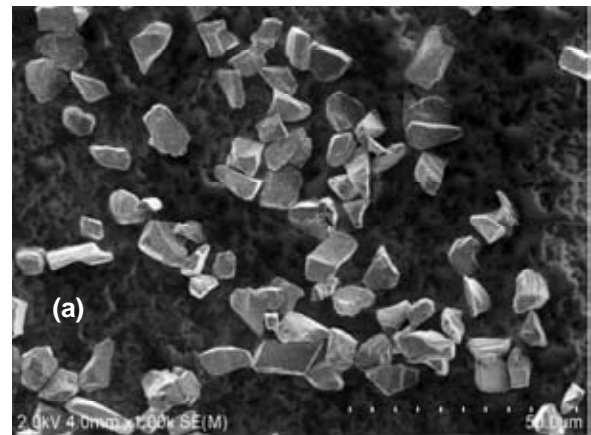


Fig. 2. Microstructure diagram of Silicon Carbide powder W_{14} by the fluidized bed under the pressure of 0.2 MPa.

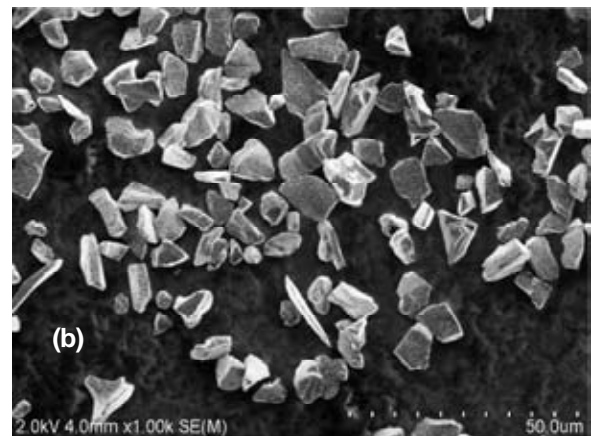
morphologies can be achieved through the same processing. Granule morphologies are different after the same processing if the dimensions of materials are different; for instance, flake material is still flake after processing generally, fibrous remains fibrous, granular remains granular. This is because the breakage usually occurs at the weak location among the chemically bonded parts. Flake minerals usually have very complete breakage. The crystals inside fibrous minerals usually grow towards the strong direction inside chemically bonded parts. Granular minerals usually have none, or three groups of cleavage [10].

Silicon carbide is an artificial abrasive made in high-temperature resistance furnace with a specific ratio of quartz, oil, coke, auxiliary wood flour, and salt. Silicon carbide is a compound of carbon atoms and silicon atoms with covalent bond. The electrons are transferred to strong bond energy SP^3 hybrid orbit when the carbon atoms and silicon atoms form the bond of each other, forming similar diamond structure with high hardness and melting point. Crystallization type of silicon carbide includes two basic crystal systems, that is, cubic silicon carbide with sphaerite structure (β -SiC), and hexagonal silicon carbide with rhombus-like structure (α -SiC).

The sharpness, breakage resistance, accumulation density and hydrophilic of abrasive ultra-fine powder are different if their shapes are transformed. As a result, the application and performance of abrasive ultra-fine powder are directly affected. For instance, the particles with sharpness angularities are easily to be carved into the machining surface than those without sharpness angularities, which results in faster and more effective grinding.



$p_1=0.5$ MPa



$p_1=0.3$ MPa

Fig. 3. Microstructure diagram of Silicon Carbide powder W_{14} by the fluidized bed under the pressure of 0.5 MPa and 0.3 MPa.

2.3. Controlling granule morphology

The fluidized-bed jet mill (LNC-3-400) was chosen as the experimental equipment in this paper. The mechanism and structure are shown in references 4 and 11 and the methods of controlling granule morphology are as follows:

(1) Controlling the grinding intensity: The collision frequency of a particle can be controlled and granule morphology can be changed to a certain extent by controlling the grinding intensity. The higher the grinding intensity is, the more qualified the particle is, and thus the more rhombus-like particles can be used for production. Otherwise, the number of superior spherical particles increases. Different nozzle diameters and the controlled grinding intensity are used to get different inlet pressures and control the outlet velocities of the nozzle. Inlet pressures will decrease when the flux through nozzles is higher than the rated flow of the air compressor. Grinding intensity can be controlled by

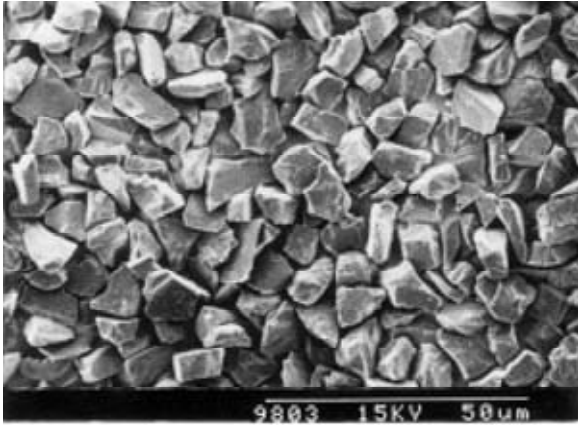


Fig. 4. Microstructure diagram of Silicon Carbide powder W_{14} by the fluidized bed with two nozzles.

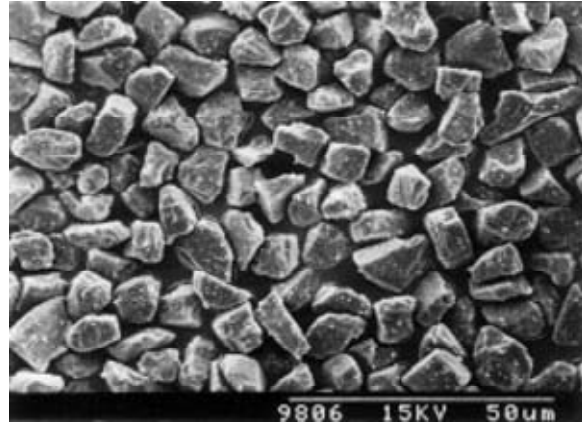


Fig. 5. Microstructure diagram of Silicon Carbide powder W_{14} by the fluidized bed with four nozzles.

changing the pressure. Comparing Fig. 2 and Fig. 3, the cracks and silicon carbide platelets become larger by increasing the pressure of grinding chamber, and more silicon carbide platelets are caused by the decrease of collision frequency. However, silicon carbide platelets decrease with the decrease of pressures in grinding chamber. Seeing from Fig. 2 and Fig. 3, diversified granule morphologies can be obtained through the fluidized bed jet mill.

(2) Different nozzle quantity: Rhombus-like and flake particles increase with the increase of grinding intensity. This is because two nozzles (opposed nozzles) are adopted in the grinding chamber of the fluidized-bed jet mill, and the collision speed and frequency of particles are increased. The particle collision frequency increases with the acceleration and collision angles of particles. The number of spherical particles increases if several nozzles (like four or five nozzles) of fluidized-bed jet mill are applied.

Grinding ultra-fine powder (W_{14}) is processed in the fluidized-bed jet mill (LNC-120-5, in a series of five classifiers) with two, three or four nozzles in the experiment [11]. As can be seen from Figs. 4, 5, and 6, the particle angularities are obviously in opposed jet mill than those in jet mill with four nozzles. The more the nozzle quantity is, the larger the spherical particles number is.

(3) Gas-solid concentration in grinding chamber: The gas-solid concentration is determined by hold up. The amount of particles present inside the mill chamber (hold up) is an important state variable as the frequency and intensity of collisions determines the grinding efficiency of the mill. Hold-ups higher or lower will result in insufficient collisions and thus result in less efficient grinding. The higher the gas-

solid concentration in grinding chamber is, the weaker the materials are accelerated by air flow. Sometimes, if the relative collision velocity of particles decreases, the amount of spherical particles increases. Otherwise, the rhombus-like particles increase with the decrease of gas-solid concentration. The gas-solid density, too high or too low, can influence the yield of the jet mill.

(4) Granularity size of particles: Each type of material and size has its own optimum hold up for obtaining the largest selectivity for breakage. The smaller the granularity size of a particle is, the more spherical particles can be obtained under the condition of the same material and the same processing. The smaller the particle size is, the less energy the particle gets, so the collision frequency of particles increases in the process of airflow grinding. On the other hand, macro and micro breakages of particles decrease and the grinding intensity of particles increases with the decrease of

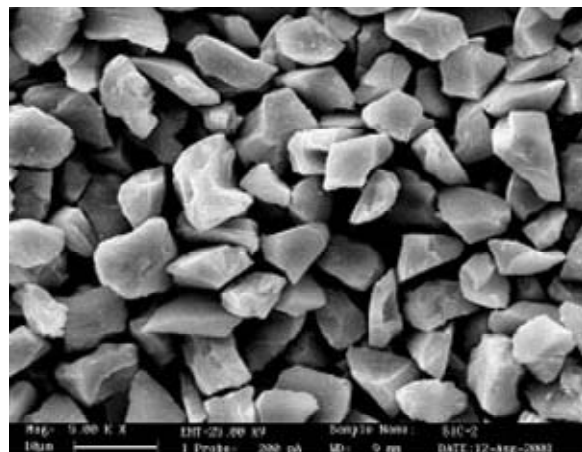


Fig. 6. Microstructure diagram of Silicon Carbide powder W_{14} by the fluidized bed with three nozzles.

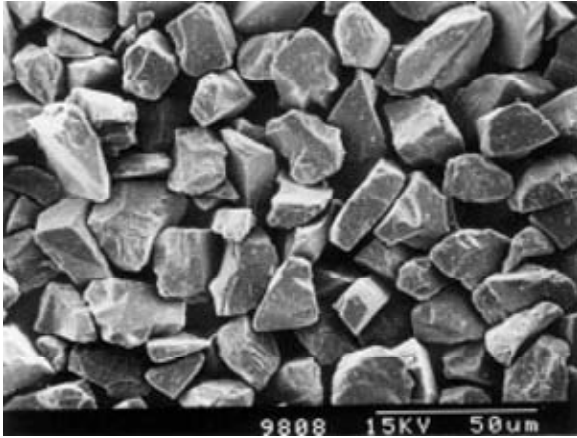


Fig. 7. Microstructure diagram of Silicon Carbide powder W_{28} by the fluidized bed with four nozzles.

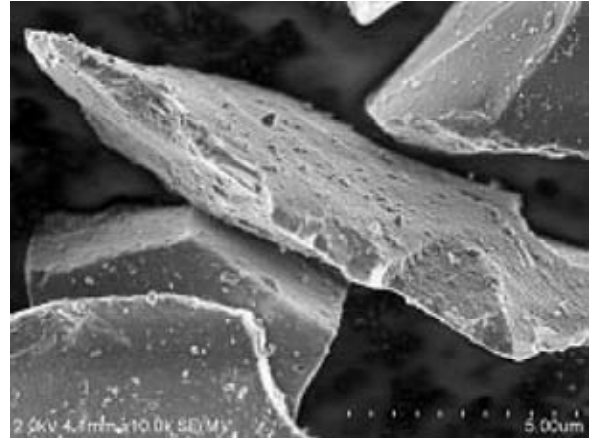


Fig. 8. Microstructure diagram of Silicon Carbide powder W_{10} by the fluidized bed in half hour.

particle size. So the collision times for grinding material increase and then the surface angularities of the particle are removed in the collision process. Comparing Fig. 5 with Fig. 7, the fluidized-bed jet mill (JZF400-5) with four nozzles was used to process ultra-fine powder like W_{14} and W_{28} . The experiment shows: the smaller the particle granularity is, the better the spherical particle is, and the shape factor is close to 1. The largest stress among particles produced by collision is generally expressed as follows:

$$\sigma_{\max} \propto m^{0.2} w^{0.4} r^{-0.6} E^{0.8}, \quad (2)$$

where m is particle quality and denoted by kg; w is particle fluidity velocity and denoted by m/s; r is curvature radius in the impact part of particle and denoted by mm; E is the elastic modulus of particle and denoted by Pa.

Curvature radius in the impact part of particles is generally larger if the particle granularity is larger, and so is the stress. Coarse particle collision and grinding intensity is required by air flow grinding. Otherwise, the required grinding intensity is lower if particle granularity is smaller. On the other hand, macro and micro cracks of particles decrease with the increase of grinding intensity for particles. The intensity increases required by grinding particles, and the materials tend to produce more collisions, making the particles remove the surface angularities in the collision process. The particles are readily grinded into eligible products if particle granularities are greater. The collision frequency of particles decreases in the jet mill process and the particle angularities increase, making a bigger shape factor.

(5) Grinding time: The influence of grinding time on particles is obvious in the grinding process, and

the collision frequency and the residence time of particles are smaller inside the jet mill if the grinding time is shorter. In regard to silicon carbide, the impact for particles is not enough if the grinding time is short as shown in Fig. 8. In this condition, the angularity is sharpness and thus meets the raw grinding. If the grinding time increases, the collision frequency of particles will increase too; therefore, the spherical granule morphology is obtained and meets the demands of precise grinding as shown in Fig. 9 and Fig. 10.

2.4. Shaping techniques for controlling granule morphology

The resulted air flow velocity cannot make particles reach the required velocity of grinding if the pressure is very low in the former chamber. The experiment shows: the key for ultra-fine shaping is to control

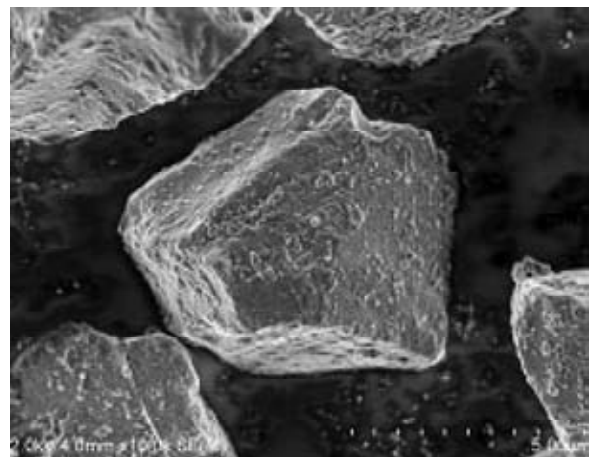


Fig. 9. Microstructure diagram of Silicon Carbide powder W_{10} by the fluidized bed in one hour.

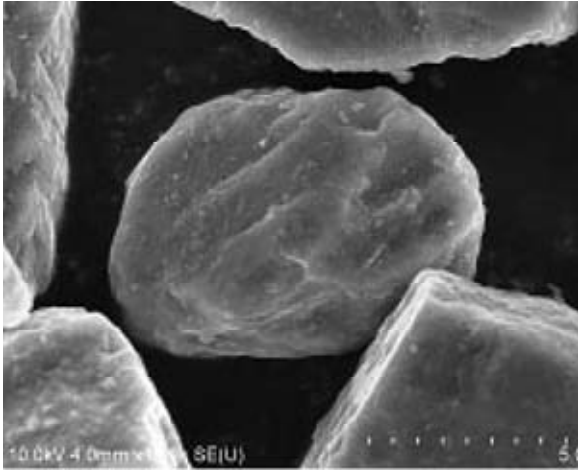


Fig. 10. Microstructure diagram of Silicon Carbide powder W_{10} by the fluidized bed in one and half hour.

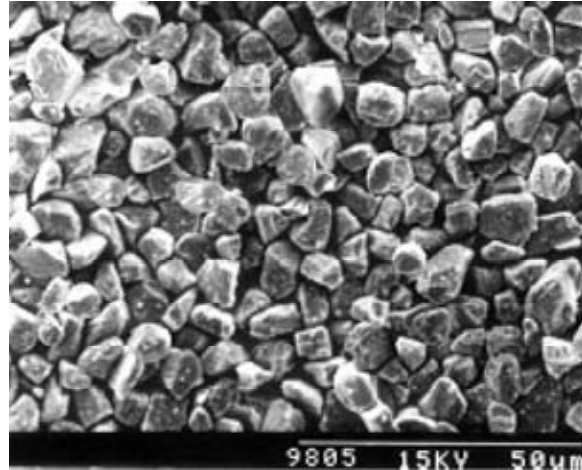


Fig. 11. Microstructure diagram of Silicon Carbide powder W_{14} by the fluidized bed with plastic.

the former chamber pressure and gas-solid concentration in the body chamber of nozzles. Energy-saving, low pressure (0.2~0.4 MPa) air compressor system and supersonic nozzles can be adopted for particle shaping of silicon carbide. The ultra-fine powder silicon carbide shown in Fig. 11, compared with that in Fig. 4, is shaping in the fluidized-bed jet mill (JNC400-2 with two air classifier in series) with four nozzles under low pressure. The whole granularities after shaping are partially fine. Compared with Fig. 5, the shape factor of silicon carbide after shaping with fine spherical particle is 1.18. This method is used to duplicate machine Chinese ink powder, which has a favorable effect.

3. PRETREATMENT TECHNIQUES FOR CONTROLLING GRANULE MORPHOLOGY

Spherical silicon carbide with uniform volume is obtained by controlling air flow grinding and air flow shaping processes. The main grinding mechanism of the jet mill is shock grinding. However, it is very difficult to control if the ideal multi-angular ultra-fine powder is to be obtained directly from the jet mill, and it is difficult to be under complete control. The research shows: a lot of cracks along weak face are produced inside the materials if the great static pressure is imposed on materials. Multi-angular (flake or sword-shaped) ultra-fine powder is readily gotten through the jet mill for dissociating or grinding materials. This high pressure roller mill is used for pretreatment equipment.

High pressure roller mill was introduced as a new type of grinding equipment in the 1980s. The large pressures (50~300 MPa) imposed on materials pro-

duce a lot of cracks inside materials, bringing about structural breakage and grinding. The surface and interior of particles are torn through high pressure roller grinding, causing plastic deformation, lattice dislocation and crack expansion in lattice. While osteoporosis material structure increases the defects, lowers the particle intensity and grinds the particle readily. The shock collision greatly increases the qualified frequency of particles at one collision after the jet mill. Therefore, the multi-angular (flake or sword-shaped) ultra-fine powder is increased. The study shows: the lower the velocity is, the bigger the roller force is when the space between two rollers is small and the roller canister surface is smooth. Therefore, the yield of high pressure roller diminishes with increased cracks and defects of particles and low particle strength. Meanwhile, rhombus-like particles increase after

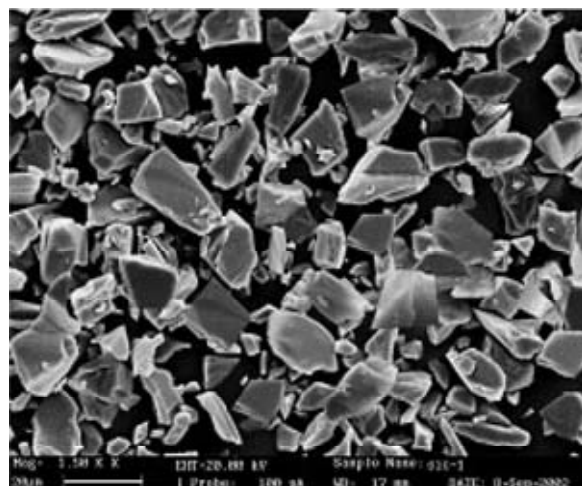


Fig. 12. Microstructure diagram of Silicon Carbide powder W_{14} by pre-processing.

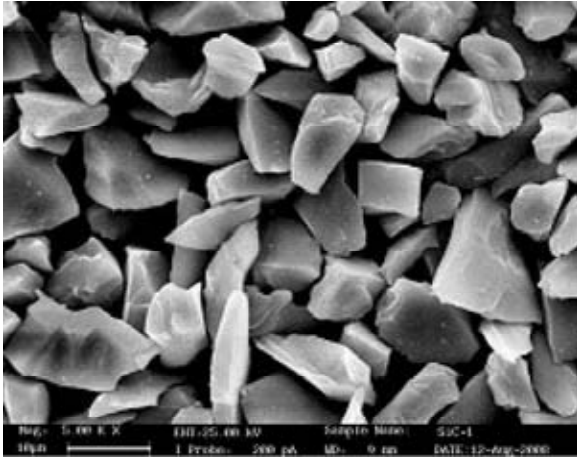


Fig. 13. Microstructure diagram of Silicon Carbide powder W_{10} by Raymond mill.

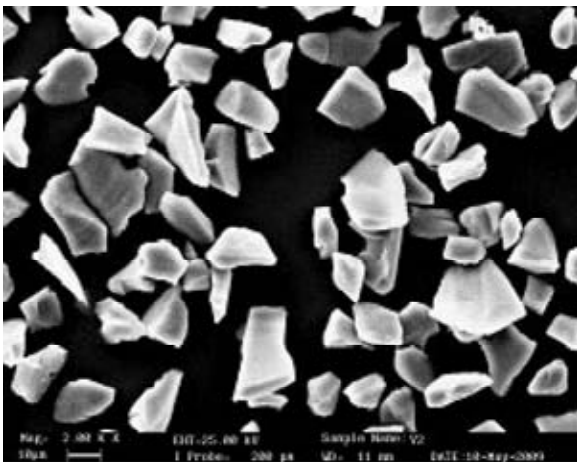


Fig. 14. Microstructure diagram of Silicon Carbide powder W_{10} by vibration mill.

dissociating or grinding. Otherwise, the rhombus-like particles decrease. The silicon carbide shown in Fig. 12 firstly goes through high pressure roller and then enters into fluidized-bed jet mill (JNC400 with one classifier) with four nozzles. Pretreatment for materials can reduce the grinding strength, increase the frequency of particle volume grinding, add the particle with multi-angular, augment particle shape factor, and enhance the grinding force of silicon carbide particles.

4 COMPARISON OF GRANULE MORPHOLOGY CONTROLLED BY JET MILL AND THE OTHER MILLS

Through the comparison, the particles prepared by jet mill under the pressure of 0.8 MPa have the uniform size distribution than those prepared by the Raymond mill shown in Fig. 13 and the vibration mill shown in Fig. 14. From the Fig. 15, the particles prepared by jet mill have fewer angularities, can be

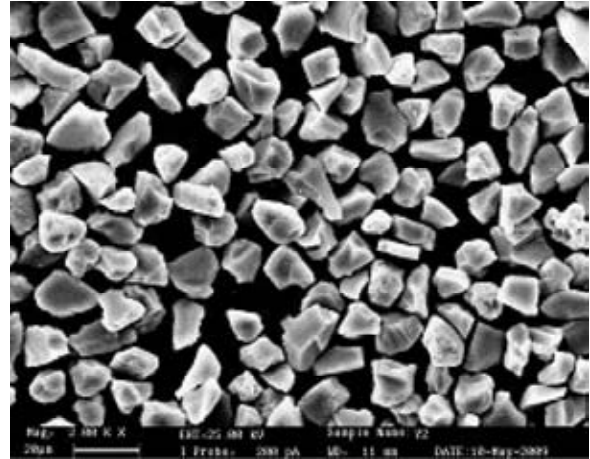


Fig. 15. Microstructure diagram of Silicon Carbide powder W_{10} by jet mill under the pressure of 0.8 MPa.

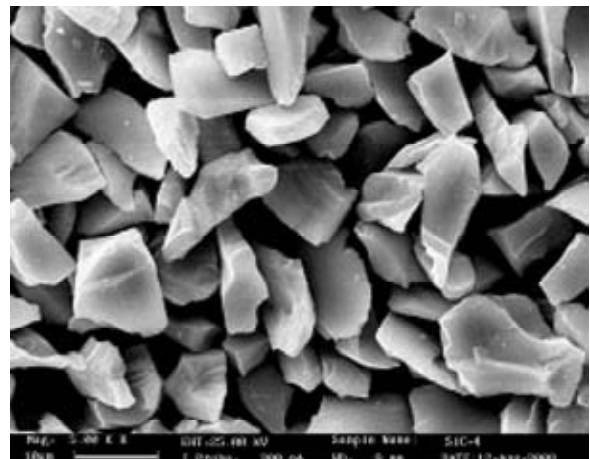


Fig. 16. Microstructure diagram of Silicon Carbide powder W_{10} by jet mill under the pressure of 1.0 MPa.

used to precise grinding of materials. However, the particles prepared by the vibration mill and Raymond mill has the more sharpness angularities, can be used to raw grinding of materials. The sharpness angularities of particulate shape can be obtained shown in the Fig. 16 through the change of flow field and pressure in improved jet mill. Under this condition, the particles have uniform size distribution and more angularities, suitable for cutting the silicon.

5. CONCLUSIONS

(1) The granule morphologies are correlated with jet mill structure (nozzle quantity), manipulated variables (comminution intensity, gas-solid concentration, pressure, grinding time), and particle properties (granular size). If these conditions are controlled to the extent, the spherical or rhombus-

like particles could also be obtained under varying experimental conditions of fluidized-bed jet milling. (2) The key for controlling granule morphology is to selectively control grinding opportunities of particles, particle interaction mechanism, collision times, intensity and residence of collisions in the process of grinding and classification. Diversified granule morphologies can be obtained by changing the jet mill structure, operation parameters and particle properties through the fluidized-bed jet mill.

(3) The high pressure roller mill is selected as pretreatment equipment. A lot of cracks along weak face are produced inside materials if the large static pressures are imposed on the materials. Multi-angular (flake or sword-shaped) ultra-fine particles are readily gotten through the jet mill for dissociating or grinding materials.

(4) Through the comparison, the particles by jet mill have the uniform size distribution than those by the vibration mill and the Raymond mill. Changing the flow field and the operation conditions is to get the required materials which meet different applications. This basic theory still needs further research.

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