A STUDY ON ULTRA FINE GRINDING OF SILICA AND TALC IN OPPOSED FLUIDIZED BED JET MILL

by

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LIST OF ABBREVIATION

Rj	rupture of joints
Cl	cleavage
Ch	chipping
Gr	ultimate grinding
ICDD	International Center for Diffraction Data
FR	Feed rate
CRS	Classifier rotational speed
GP	Grinding pressure
GCP	Grinding chamber pressure

LIST OF SYMBOLS

P	Grinding pressure
N	Classifier rotational speed
ρ	density
B	half width
θ	the Bragg angle of (*h k l*) reflection
ε	lattice strain
D	crystallite size
C	crystallinity index
B_o	background of the feed
B	background of the ground samples
I_o	integral intensities of diffraction lines for feed
I	integral intensities of diffraction lines for ground samples
Am	amorphous fraction
A_o	areas under the prominent plane for feed
A	areas under the prominent plane for ground sample
H_h	height of holdup
H_c	the distance between surface of holdup and classifier
x	distance between top of grinding-classifying chamber and surface of holdup
$\bar{x}$	average value
n	number of samples
x_i	data collected
d(4.3)	Volume moment diameter
d_{50}	Mean particle size
x_k	number percentage of detected diameter
d_k	detected diameter

ψ	span values
d_i	i% smaller than
K_α	spectral line for an element
β_f	integral breadths of the instrumental
β_h	integral breadths observed
β_g	integral breadths of measured profile
D_v	volume weighted mean of the crystallite size
K	constant
θ	Bragg angle of (*h k l*) reflection
λ	wavelength of X-rays used
ε	lattice strain
r	rotor radius
v_φ	circumferential velocity
v_r	radial velocity
x_c	cut size
ρ_s	density of solids
η	dynamics viscosity
$R_{max}^{(4.3)}$	maximum reduction ratio
$d_{4.3}^{f}$	volume moment diameter of feed
$d_{4.3}^{p}$	volume moment diameter of ground product
$\mathbf{Abs}_{n}$	Absorption at wavelength *n*
$E_{kinetic}$	kinetic energy
m_g	mass of gas
v	gas velocity
φ	gas constant
P	gas pressure

ρ_g	Gas density
SEC	specific energy consumption
F	feed rate
C	Circularity
D_{eq}	Equivelent diameter
A	Area
P	Perimeter

LIST OF PUBLICATION

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Samayamutthirian Palaniandy, Khairun Azizi Mohd Azizli, Mariatti Jaafar, Farrah Noor Ahmad, Hashim Hussin and Syed Fuad Saiyid Hashim. (2007) Effect of structural changes of silica filler on the coefficient of thermal expansion (CTE) of underfill encapsulant. *Powder Technology*, *In Press, Accepted Manuscript*.

F.N. Ahmad, Mustapha Mariatti, Samayamutthirian Palaniandy and Khairun Azizi Mohd Azizli. (2007). Effect of particle shape of silica mineral on the properties of epoxy composites. *Composites Science and Technology*, *In Press, Corrected Proof*.

Samayamutthirian Palaniandy, Syed Fuad Saiyid Hashim, Hashim Hussin and Khairun Azizi Mohd Azizli. (2004). Structural Defects during Fine grinding Process. *Journal of Solid State Science and Technology Letters* .11, 2 (Supplementary).

Samayamutthirian Palaniandy, Khairun Azizi Mohd Azizli Syed Fuad Saiyid Hashim, and Hashim Hussin. Microstructure characterization of mechanically activated nanocrystallite quartz using XRD line broadening. (2006) Proceeding of International

Conference on X-Ray and Related Techniques in Research and Industry. Penang, Malaysia. Putrajaya, Malaysia.

Samayamutthirian Palaniandy, Syed Fuad Saiyid Hashim, Hashim Hussin and Khairun Azizi Mohd Azizli. (2004). Effect of hardness on the particle morphology during fine grinding process in jet mill. *13th Scientific Conference Electron Microscopy Society Of Malaysia.* Putrajaya, Malaysia.

Samayamutthirian Palaniandy, Khairun Azizi Mohd Azizli Syed Fuad Saiyid Hashim, and Hashim Hussin. Effect of grinding pressure and classifier frequency on the particle shape of silica ground in Opposed fluidized be jet mill. (2006) *15th Scientific Conference Electron Microscopy Society Of Malaysia.* Penang, Malaysia.

Samayamutthirian Palaniandy, Khairun Azizi Mohd Azizli Syed Fuad Saiyid Hashim, and Hashim Hussin. Effect of Jet Mill's Operational Parameters on the product quality during Silica Grinding. (2007). Proceeding of PSU-UNS International Conference on Engineering and Environment, Phuket, Thailand.

Samayamutthirian Palaniandy, Khairun Azizi Mohd Azizli Syed Fuad Saiyid Hashim, and Hashim Hussin. Microstructure Characterization of Mechanically Activated Talc in Jet Mill using Line Broadening Technique. (2007) Proceeding of International Conference on Advancement of Materials and Nanotechnology. Langkawi, Malaysia.

A Study on Ultra Fine Grinding of Silica and Talc
In Opposed Fluidized Bed Jet Mill

ABSTRACT

Fine grinding is normally carried out in energy intensive grinding mills such as jet mill. Besides size reduction, jet mill also induced mechanochemical effect on the ground product. Fine grinding testwork of silica and talc was carried out in jet mill by varying the feed rate, classifier rotational speed and grinding pressure at five levels. In this jet mill, grinding and classification took place simultaneously. The grinding chamber pressure, holdup height and mass, classifier current and specific energy consumption were determined. The ground products were characterized in terms of particle size distribution and mechanochemical effect via X-ray diffraction and Infrared Spectroscopy (IR). The ground product exhibit poly-modal distribution so its particle size was characterized by volume moment diameter. Coarser particle size distribution was obtained at higher classifier rotational speed due to the fluctuation of inside mill condition and particles characteristics which deviated from Stokes law. The optimum feed rate, classifier rotational speed and grinding pressure for silica and talc were 4 kg/h, 13000 rpm, 5 bars and 8 kg/h, 7000 rpm, 3 bars respectively. The optimum holdup mass ranged from 200g to 400g. The optimum holdup height and distance between the holdup surface and classifier were 10 cm and 12.5 cm respectively. Under pressure condition in grinding-classifying chamber produced finer particles. At lower grinding pressure, abrasion breakage mechanism is dominant for silica whilst delamination of nano-layer was dominant for talc. At high grinding pressure, silica and talc exhibited destructive breakage mechanism. Ground silica and talc exhibited reduction in circularity values. Ground silica and talc exhibited mechanochemical effect where the degree of crystallinity ranged from 71.1% to 90.9% and 43.3% to 85.3% respectively. The crystalline size and lattice strain of silica ranged from 22.71 nm to 35.54 nm and 0.024 to 0.037 respectively. The crystalline size and lattice strain of talc ranged from 147.69 nm to 353.72 nm and 0.08 to 0.2 respectively. Reduction of hydroxyl band and broadening of Si-O stretching band for silica and Si-O-Mg and hydroxyl band for talc were observed via IR. Optimum specific energy consumption was 100kWh/ton and 1000 kWh/ton for silica and talc respectively.

Kajian Tentang Pengisaran Ultra Halus Silika Dan Talkum Di Dalam Pengisar Jet Lapisan Bendalir Bertentangan

ABSTRAK

Proses pengisaran halus biasanya dilakukan di dalam pengisar intensif tenaga seperti pengisar jet. Selain daripada pengurangan saiz, pengisar jet juga meghasilkan kesan mekanokimia terhadap partikel yang dikisar. Kajian pengisaran halus silika dan talkum telah dilakukan di dalam pengisar jet dengan mengubah kadar suapan, halaju putaran pengkelas dan tekanan pengisar pada lima peringkat. Proses pengisaran dan pengkelasan berlaku serentak di dalam pengisar jet ini. Tekanan dandang pengisar, jisim dan ketinggian lapisan bendalir, arus pengkelas dan penggunaan tenaga spesifik telah ditentukan. Produk terkisar dicirikan berdasarkan taburan saiz partikel dan kesan mekanokimia menggunakan Belauan Sinar-X dan Spektrskopi Inframerah. Produk terkisar menunjukkan taburan saiz partikel poli-model maka saiz partikel dicirikan dengan diameter momen isipadu. Saiz partikel yang kasar diperolehi pada halaju putaran pengkelas yang tinggi disebabkan perubahan keadaan di dalam pengisar dan ciri partikel bercanggah dengan Hukum Stoke. Kadar suapan, halaju putaran pengisar dan tekanan pengisaran yang optimum untuk silika dan talkum masing-masing adalah 4 kg/h, 13000 rpm, 5 bar dan 8 kg/h, 7000 rpm, 3 bar. Jisim lapisan terbendalir optimum adalah diantara 200g hingga 400g. Ketinggian optimum lapisan terbendalir dan jarak optimum antara lapisan terbendalir dan pengkelas adalah masing-masing 10 cm dan 12.5 cm. Keadaan tekanan bawahan menghasilkan produk yang lebih halus. Pada tekanan pengisaran rendah, mekanisme pemecahan lelasan adalah dominan untuk silika manakala dilaminasi lapisan-nano pula dominan untuk talkum. Pada tekanan pengisaran yang tinggi, silika dan talkum mengalami mekanisme pemecahan membinasa. Silika dan talkum terkisar menunjukkan pengurangan nilai indeks kebulatan. Silika dan talkum terkisar mengalami kesan mekanokimia di mana darjah pengkristalan adalah masing-masing diantara 71.1% sehingga 90.9% dan 43.3% hingga 85.3%. Saiz kristal dan terikan kekisi silika masing-masing di dalam julat diantara 22.71 nm hingga 35.54 nm dan 0.024 hingga 0.037. Manakala, saiz kristal dan terikan kekisi talkum masing-masing dalam julat diantara 147.69 nm hingga 353.72 nm dan 0.08 hingga 0.2. Pengurangan jalur hidroksil dan pengembangan jalur regangan Si-O untuk silika dan jalur Si-O-Mg dan hidroksil untuk talkum diperhatikan melalui FTIR. Penggunaan tenaga spesifik optimum masing-masing untuk silika dan talkum adalah 100 kWjam/tan dan 1000 kWjam/tan.

CHAPTER 1

1.0 Introduction

The scope of grinding can be divided into three categories which are coarse grinding, fine grinding and mechanical activation and is distinct by the amount of energy delivered by the grinding mills to the grinding tools and the materials to be ground (Boldyrev et. al., 1996; Tkacova, 1989). The objective of coarse grinding is for size reduction while mechanical activation is for structural changes of the ground particles to increase the reactivity of the particles. Fine grinding is an intermediate case between coarse grinding and mechanical activation and is gaining its importance as the demand for fine particles from various industries such as paper, paint, plastic, pharmaceuticals, ceramics, cosmetics, foods and fine chemicals is increasing drastically (Belaroui *et. al.*, 1999; Belaroui *et. al.*, 2002; Molina-Boisseau *et. al.*, 2002; He *et. al.*, 2004; Frances *et. al.*, 1996). These industries demand very stringent fine particle specifications in terms of particle size and its distribution (Belaroui *et. al.*, 2002). Besides these two criteria, lately the degree of crystallinity of fine mineral particles below 10 µm is becoming more important as fine grinding process produces particles with nanocrystallite structure which exhibits abnormal behavior such as enhance hydrometallurgy process, low annealing temperature, reactivity improvement of cementitious materials, formation of metastable phases and particles with high surface energy (Tkacova, 1989). Table 1.1 shows the characteristics and application of silica and talc as mineral fillers by various industries.

Fine grinding is normally carried out in high intensity grinding mills such as planetary mill, attrition mill, oscillating mill and jet mill. These mills deliver huge amount of energy for particle breakage to produce particles below 10µm. However, mechanochemical effect is induced by fine grinding process when the power bulk density is more than 0.1 kWm^{-3} (Tkacova, 1989). The severe and high intensity energy delivered by the grinding mill onto the particles leads to structural changes near surface region where the solids come into contact under mechanical forces besides size reduction. The structural changes induces changes in crystallinity, crystallite size and lattice strain. These changes are generally termed as mechanochemical effect (Venkataraman and Narayanan, 1998). Vibration mill, stirred mill, jet mill and planetary mill experienced mechanochemical effect during the fine grinding process, which agrees with Table 1.2.

Table 1.1: Characterization of silica and talc as mineral fillers in various industries (Jones, 2002; O'Driscoll, 1992; Loughbrough, 1993)

Industries	Minerals	d_{50}, μm	Function	Phase
Plastic	Silica	2.5-13	Filler/extender/ reinforcement	Crystalline/ amorphous
	Talc	1-20	Filler	
Adhesives /Sealant	Silica	0.05-2	Filler	amorphous
	Talc	10	Filler/Extender	NA
Paint	Silica	2.5-10	Filler	Crystalline
	Talc	1.7-9	Filler/Extender	Crystalline
Paper	Talc	5	Filler	Crystalline
Cosmetics	Talc	5	Filler/Lubricant	NA
Pharmaceutical	Talc	1	Filler/dusting	Crystalline/ amorphous

NA – Not available

Table 1.2: Power bulk densities in various grinding mills (Tkacova, 1989)

Grinding mills	Power bulk densities (kWm^{-3})
Ball mill	0.01 - 0.03
Peripheral mill	0.03 – 0.10
Vibration mill	0.03 – 0.90
Jet mill	0.07 – 2.00
Stirred mill	0.10 – 1.00
Planetary mill	0.50 – 5.00

Although fine grinding will induce mechanochemical effect that enhances certain properties of the downstream industries, but certain application needs crystalline particles (Samtani et. al., 2001). The mechanochemical effect can't be avoided during fine grinding process as it happens concurrently with the size reduction process (Begin-Colin *et. al*., 2000). The challenge is to control the energy intensity induced on the particles so that the ground product can maintain its crystallinity if needed. The control of partially amorphous and crystalline particles at certain percentages to increase the particle reactivity without jeopardizing the quality will be the future for the production of fine particles.

1.1 Mechanochemical Effect Induced By Fine Grinding Process

During mechanical stressing in fine grinding process, part of the energy is stored in the materials (Iguchi and Senna, 1985). In the case of nonmetallic minerals which have low thermal conductivity, the energy delivered by the grinding mills is not stored in the particles as thermal energy but applied to bending or breaking of crystal. In fine grinding process, apart from producing important changes in the properties of the solid, leads to structural alteration by loss of regularity in the crystalline network (amorphization) as well as formation of active surfaces and leads to increase reactivity as shown in Figure 1.1 (Aglietti *et. al.*, 1986a; Al-Wakeel, 2005; Tkacova, 1989). According to Juhasz and Opoczky (1990), 3.7% of the total energy supplied to a vibration mill during quartz grinding was used for phase transformation while the major portions were lost as heat and mechanical losses.

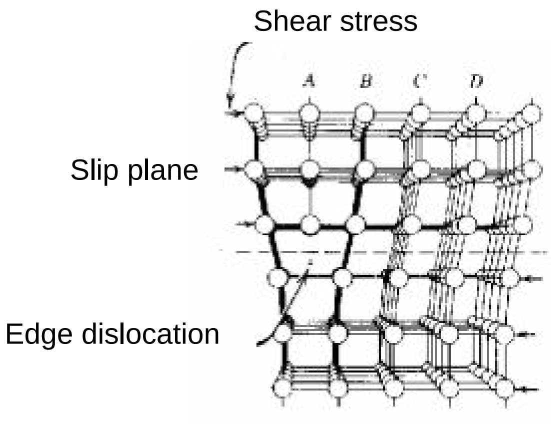

Figure 1.1: Loss of regularity in the crystalline network (Tkacova, 1989)

Mechanochemical effect in fine particles has created interest among researchers due to the abnormal behavior of the ground fine particles which has crystallite size in nanometer range (Bid *et. al.*, 2001). These particles exhibit abnormalities such as increase in reactivity, reduction in initial thermal decomposition temperature, phase transformation, decrease in sintering temperatures and enhancement of leaching process due to amorphization and smaller crystallite size. (Gonzalez et.al., 2000; Zhang *et. al.*, 1996; Temuujin *et. al.*, 1999; Begin-Colin *et. al.*, 1999; Sanchez *et. al.*, 2004; Mi *et. al.*, 1999; Welham, 1998; Kalinkin *et. al.*, 2003; Ryou, 2004; Hu *et. al.*, 2003; Benezet and Benhassaine, 1999).

Another issue that relates to mechanochemical effect during fine grinding process is the existing of grinding limit. Boldyrev *et. al.* (1996) mentioned that there was a certain size called tough-brittle transition size where particle above this size particle would experienced brittle breakage whilst particles smaller than this size would experienced plastic deformation. The stress required to break a small particle may be rather high, especially if the material is hard. The mechanical impacts in the grinding mill were obvious to be limited by intensity which was determined by mill design and the operating condition (Boldyrev *et. al.*, 1996). Mechanochemical effect took place if the particle size was smaller than the tough-brittle transition size, and secondly, the stress on the particle should be higher than yield stress, which depended on the particle size (Boldyrev *et. al.*, 1996).

Ground product produced through fine grinding process exhibited high surface energy. The increased in the surface energy on the ground particles led to a major problem which was interaction between fine particles which resulted in the formation of larger particles especially when the particle size was less than 10 µm. Juhasz and Opoczky (1990) had suggested that there were three stages of interaction between the particles which were adherence, aggregation and agglomeration. At adherence stage, particles would coat the apparatus and the grinding bodies. At aggregation stage, the particles were associated by weak van der Waals type of adhesion and it was reversible interaction. Agglomeration was defined as a very compact, irreversible interaction of particles in which chemical bonding may also play a role which may occur during fine grinding process. Agglomeration is detrimental to both the activity and the quality of the product. Currently, grinding aids are used to avoid the agglomeration during fine grinding process (Godet-Morand *et. al.*, 2002; Rajendran and Paramasivam, 1999; Frances *et. al.*, 1996).

1.1.1 Mechanochemical Effect Induced In Jet Mill

Jet mill is classified as a high intensity grinding mill which is normally used to produce particles below 10 µm (Alfano *et. al.*, 1996; Eskin and Voropayev, 2001; Tasirin and Geldart, 1999; Benz *et. al.*, 1996). It is widely used in mineral, pharmaceutical and fine chemicals industries compared to other types of size reduction machines due to its narrow size distribution as obtained by Choi *et. al.* (2004), the absence of contamination due to autogeneous grinding mechanism, low wear rate, small footprint, low noise and the ability of grinding heat sensitive materials (Berthiaux and Dodds, 1999; Midoux *et. al.*, 1999; Kolacz, 2004; Muller *et. al.*, 1996).

Mechanochemical effect normally takes place when batch grinding is carried out for longer grinding period in high intensity grinding mills (Kanno, 1985; Gonzalez *et. al.*, 2000). Jet mill is a continuous type grinding mill with internal classification which means size reduction and classification will take place simultaneously. In jet mill the retention time of particles in the grinding chamber is very short because the particles will be classified out as soon as it reaches the desired size.

Not much attention is given on the mechanochemical effect in continuous grinding process especially when the grinding and classification process occur simultaneously in short grinding period. Although in jet mill the grinding and classification process occurs simultaneously and the retention time of the particles is believed to be very short, but the energy intensity delivered to the particle is huge which may result in mechanochemical effect in short grinding period. Chung *et. al.* (2003) and Choi *et. al.* (2004) reported that significant structural changes did not take place when ursodeoxycholic acid was ground in jet mill where the reduction of crystallinity was only 2%. Contradicting to Chung *et. al.* (2003) and Choi *et. al.* (2004), Juhacz and Opoczky (1990) reported that cement clinker, dolomite, limestone and silica ground in jet mill showed the highest reactivity due to the feed which suffered deep-seated changes in its crystal structure induced by high velocity impact.

1.2 Problem Statement

Besides size reduction, fine grinding process induces mechanochemical effect that changes physicochemical characteristics of particles in energy intensive grinding mills (Venkataraman and Narayanan, 1998). The amount of energy delivered to the particles and breakage mechanism controls the intensity of mechanochemical effect of particles ground in jet mill. These two factors are controlled by the operating parameters and inside mill conditions of the jet mill. The inside mill conditions such as grinding chamber pressure and amount of holdup in the grinding chamber which will influence the breakage mechanism are controlled by the operating parameters such as feed rate, classifier frequency and grinding pressure.

Jet mill is operated through autogeneous grinding process where the particle breakage is due to high velocity inter-particles impact collision. The autogeneous grinding promotes particle breakage along the weak planes thus producing stronger fine particles with smooth surfaces. The attrition process between the particles in the grinding chamber removes the sharp edges especially for brittle particles producing more spherical particles.

The breakage mechanism is an important factor in determining the intensity of mechanochemical effect in ground particles. The shear and impact breakage mechanism is believed to contribute to the mechanochemical effect in the ground product in jet mill. The breakage mechanism in the jet mill is very much dependent on the operational condition of the jet mill which controls the impact or shear breakage intensity and also the retention time of the particles in the grinding chamber. Grinding chamber pressure plays an important role in determining the retention time of the particles in the grinding chamber. The amount of holdup in the grinding chamber is essential to obtain successive particle breakage which later leads to mechanochemical effect. The types of breakage mechanism which is either abrasion or destructive will be determined by the grinding pressure where by at high grinding pressure impact breakage is more dominant whilst at low grinding pressure shear breakage is prominent (Berthiaux and Dodds, 1999). The degree of crystallinity, crystallite size and lattice strain of the ground particles are directly related to the intensity of mechanochemical effect, which is actually controlled by the operating condition of jet mill. Thus, the study on the control of the operating parameters and inside mill conditions is very essential to optimize the size reduction process and mechanochemical effect of the ground particles.

Silica and talc have great industrial application in the form of fine particles especially when the particle size is below 10µm. Silica is widely used as filler in plastic, adhesive, paint whilst talc applications as filler is in plastic, paint, paper, cosmetics and pharmaceutical. These industries have set a stringent specification for the filler which includes particle size and its distribution, particle shape and degree of crystallinity (Sanchez et. al., 2004).

1.3 Objective

The main objective of this research work is to study the size reduction process and mechanochemical effect induced in jet mill during fine grinding process of silica and talc and the influence of operating parameters and breakage mechanism. The measurable objectives of this study are:

1. To determine the effect of operating conditions such as feed rate, grinding pressure and classifier rotational speed on the grinding chamber pressure and amount of holdup in the grinding chamber.
2. To determine the breakage mechanism in the grinding chamber according to the operating conditions of the jet mill.
3. To study on mechanochemical effect of silica and talc at various operating conditions which includes determination of degree of amorphism, crystallite size and lattice stain of the ground product.
4. To study the specific energy consumption of jet mill.

1.4 Scope Of Work

The scope of this work includes fine grinding process of silica and talc in jet mill by varying the operational parameters such as feed rate, classifier rotational speed and grinding pressure. The specific kinetic at each operating parameters will be determined. The inside mill condition such as amount of holdup and grinding chamber pressure will be studied. The breakage mechanism of the particles at different operating parameters will be studied as well. The influence of breakage mechanism and specific kinetic energy on the mechanochemical effect of products will be determined. The outcomes of this research work will provide a fundamental understanding on the effect of breakage mechanism on mechanochemical effect of ground product and therefore optimizing the size reduction process in the jet mill.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

Fine mineral particles ranging from 1μm to 10μm play an important role as fillers in various industries such as plastics, paint, paper, sealant and adhesives, cosmetics, detergent, ceramics, agriculture and pharmaceutical (Benz *et al*., 1996). Normally fine mineral particles is produced through grinding process in various energy intensive grinding mills such as oscillating mill, planetary mill, vibration mill, stirred mill and jet mill.

Fine grinding process is defined as production of fine particles in the size range of several micrometers (Benz *et al.* 1996). Tkacova (1989) mentioned that energy intensive mills are often used for ultra fine grinding and mechanical activation purposes due to the huge amount of energy delivered to the particles that were being ground.

Although the main objective of fine grinding process is size reduction in several micrometer range, there are other phenomenon that take place during this process due to the amount of energy delivered. Severe and intense mechanical actions on solid surfaces are known to lead to physical and chemical changes in the near-surface region where the solid come into contact under mechanical forces. These mechanically initiated chemical and physicochemical effects in solids are generally termed as mechanochemical effects (Venkataraman and Narayanan, 1998).

As fine grinding process is an intermediate process between size reduction and mechanical activation hence the amount of energy delivered to the particles during fine grinding process will dictate the intensity of mechanochemical effect on the ground particles and it takes place simultaneously during the fine grinding process (Boldyrev et. al., 1996).

Known for its high energy consumption and inefficiency, fine grinding of materials has been long looked at as an area with huge scope of development. It consumes a major portion of the power drawn from a typical mineral processing plant and consumes approximately 3% of the electricity generated in the industrialized country of the world (Ipek *et al.,* 2005). Fine grinding is normally performed to produce particles below 10μm as its demand is becoming very important in various fields. These reasons have motivated researchers around the world to achieve optimum energy utilization with substantial economic benefits. Currently, these fine particles are very expensive and the production rates are relatively small. It is not easy to meet the stringent demand on its fineness, purity, shape and crystallinity due to the following reasons:

- The particle strength increases strongly with decreasing particle size, so the particle breakage intensity has to be high.
- Particles of brittle materials deform plastically below the tough brittle transition size which depends on the types of materials. The breakage of this particle becomes difficult (Boldyrev *et al.*, 1996).
- The adhesion force in the fine particles causes agglomeration.

Among the industrial minerals, fine particles of silica and talc are vastly used as mineral fillers in various industries. Silica and talc have common use as mineral fillers in paint, plastics, ceramics and agriculture industries. Silica is largely consume in sealant and adhesives, glass, building application, chemicals and metallurgical industries, whilst talc is used in paper, cosmetics, detergent and pharmaceutical.

Micronized silica used as extender and filler in the paint industry will increase the resistance to chemical attack due to its acid resistance, whilst its hardness improve scrub ability and burnish resistance. Its narrow size distribution and bright colour minimize the resin demands and improve colour acceptance. Silica is chosen as mineral filler due to its heat resistance, low thermal expansion, chemical resistance and high dielectric strength. Its inertness and durability enhances the physical strength of large epoxy casting for example laboratory bench top (Moore, 2002).

Talc is used as filler, to control pitch and stickies and in coating formulation in paper industry. The advantages of talc as filler are improve smoothness, porosity, opacity, abrasion and yellow index. Talc is among the most common extender used in paints due to its platy shape can reduce permeability. The function of talc in paint is as reinforcement, reduce sagging and allow it to flatten out resulting smooth film. This property has enhanced the consumption of talc in domestic, industrial and protective coating. Ultra fine talc is used in plastic to improve its mechanical and surface properties such as stretch resistance. Talc is loaded in plastic ranging from 20% to 40% to enhance stiffness, tensile strength and creep resistance. The optimum particle size for this application is 8µm. Another talc application of talc in the plastic industry is as antiblocking agent for films (Russell, 1989).

Most of these applications need fine particles below 10µm with narrow size distribution. The stringent demand from the industries according to the specific application has made the grinding industry to move ahead to produce custom made mineral fillers that specially can suit for the specific industrial product application. Table 2.1 shows the industrial demand for various mineral fillers used in product manufacturing (Russell, 1989).

Table 2.1: Key physical properties of selected mineral fillers (Russell, 1989)

Mineral	d_{50} (μm)	Surface area, (m^2/g)	Specific gravity (g/m^3)	Aspect ratio	Mohs' hardness
Ground silica	2-6	1-2	2.65	Low	7.5
Talc	1-1.5	6-10	2.8	Moderate	1.5

2.2 Energy Intensive Grinding Mills

High intensity grinding mills such as planetary mill, stirred mill, jet mill, vibration mill and peripheral mill could exhibit mechanochemical effect due to the high power bulk density compared to ball mill (Tkacova, 1989). These mills are normally used for fine grinding or mechanical activation process. These mills are chosen for fine grinding process as they have huge amount of energy that can be use for fine particle breakage and the excessive energy will be used for crystal structural changes besides transformation to heat energy.

Grinding mills can be divided into three main groups according to the method of energy transfer from mill to ground material. Kinetic energy from the mill can be transmitted as follows (Juhasz and Opoczky, 1990):

- to mill body, from where it is transferred to the grinding media and charge to be ground by friction, centrifugal and gravitational force such as ball mill.
- directly to crushing elements such as peripheral mill and roller mills.
- directly to material being ground by means of a flowing stream of the carrier gas which creates vigorous collision between particles and their impingement on a rigid obstacle such as jet mill.

Various types of energy intensive mill in which, the energy density is several times greater than ball mill, is often used for fine grinding. These types of mill can be grouped into two categories, non-rotary ball or beads mill and impact mill. The non-rotary mills are planetary, vibration and stirred mill. Jet mill is an impact type mill.

The grinding intensity in these fine grinding mills is based on acceleration and predominant mode of stress (Tkacova, 1989). The acceleration of the vibration mill is specified by its frequency and amplitude. The industrial vibration mill which operates at frequencies of 16-19 revolution s^{-1} and amplitudes below 6 mm accelerates ten times greater than gravitational acceleration. Besides acceleration, density and shape of grinding media, continuous operations and the filling rate also affect the fine grinding process (Juhasz and Opoczky, 1990). Grinding and activation effect increase with the increase of grinding media density in vibration mill. The materials in the vibration mills are predominantly stress by compressive and impact mode pulses imparted by the grinding media. Attrition particle breakage mode is effective between the media with different speeds and between the media and drum shell.

Planetary mill is relatively new but a promising equipment due to its high energy bulk density. The enhancement of acceleration in relation to conventional ball mill is achieved by the combined action of two centrifugal fields. Laboratory and pilot plant planetary mill attain acceleration which may exceed gravitational acceleration by a factor of 100 (Juhacz and Opoczky, 1990). The ultra fine grinding and mechanical activation in this mill is very efficient. The breakage mechanism in planetary mill depends on the motion of the grinding media. The modes of grinding media movement in planetary mill are cascading, cataracting and hurricane (Juhacz and Opoczky, 1990).

Special types of non-rotary bead mill include the stirred mills. In stirred mill, the material is comminuted by means of free flowing beads which are set in motion by a stirrer. The grinding effect depends on the stirrer speed, chamber geometry, rheological properties of slurry, bead size and types and amount of charge. Stirred mill can be used for grinding soft materials such as soft pigments and also very hard ceramics as the operating condition and design can be varied according to material properties. The breakage mechanism in stirred mill is through shear supplemented with compression.

Two types of impact type mills used in fine grinding are high peripheral speed mill and jet mill. In high peripheral speed mill, two wheels rotate with different speeds, usually in the opposite direction thus doubling the shock velocity. Pin breakers are situated on the rotor along the concentric rows of circle. Materials are centrally fed and when it moves from the center to the periphery of rotor, individual particles collide with pin breakers and each other. The mode of stress is determined by relative circumferential velocity of rotor. The relative circumferential velocity may be as high as 200ms^{-1} to 300 ms^{-1} (Juhacz and Opoczky, 1990).

2.3 Jet mill

Jet mill has gain its importance in producing high purity fine particles ranging from 1μm to 10μm since it was first invented in 1960 (Midoux *et al,* 1999). There are two common types of jet mill which are the spiral jet mill and the fluidized bed jet mill. The advantages of jet mill in producing fine particles are high fineness combines with narrow size distribution, high degree of dispersion, low grinding chamber temperature, no agitated built in elements, high turbulence in the grinding chamber, which means high heat transmission and high mass transfer with no product contamination through wear (Muller *et al*, 1996).

Although jet mill has several advantages but it is still an energy intensive process as only 2% - 5% of the energy supplied is used to create new surfaces (Mebtoul *et al.,* 1996; Alfano *et al.*, 1996; Lecoq *et al.,* 2003). Gommeren *et al.* (2000) had carried out the modelling of the jet mill plant to reduce the energy by 50%. The development of more energy efficient fine grinding process in jet mill implies a better fundamental understanding of the various mechanisms involved in the fragmentation of the particles such as grinding chamber pressure, amount of holdup in the grinding chamber, particle shape, air flow in the grinding and classification chamber besides the operational parameters and design parameters such as, classifier speed, nozzle distance, feed rate and grinding pressure (Kolacz, 2004).

Figure 2.1 shows the schematic diagram of opposed fluidized bed jet mill. The fluidized-bed jet mill consists of a vertical grinding chamber with opposing gas jets near its lower end. Particles contained in the hopper is continuously fed by screw feeder into the grinding chamber where they are ground at the meeting point of the three concurrent air jets formed by ceramic nozzles supplied with compressed air. The breakage mechanism in the grinding chamber is primarily due to impact and abrasion with other particles and very much dependent on the operating condition of the jet mill. After grinding, the particles are carried out by air flow up to the wheel classifier, which removes the fine particles and return the coarser ones to the grinding zone resulting in relatively tight particle-size distributions (Godet-Morand, 2002). Figure 2.2 shows the flow of the particles in the grinding-classifying chamber. The fine particles are collected in a container below an air cyclone (Berthiaux and Dodds, 1999).

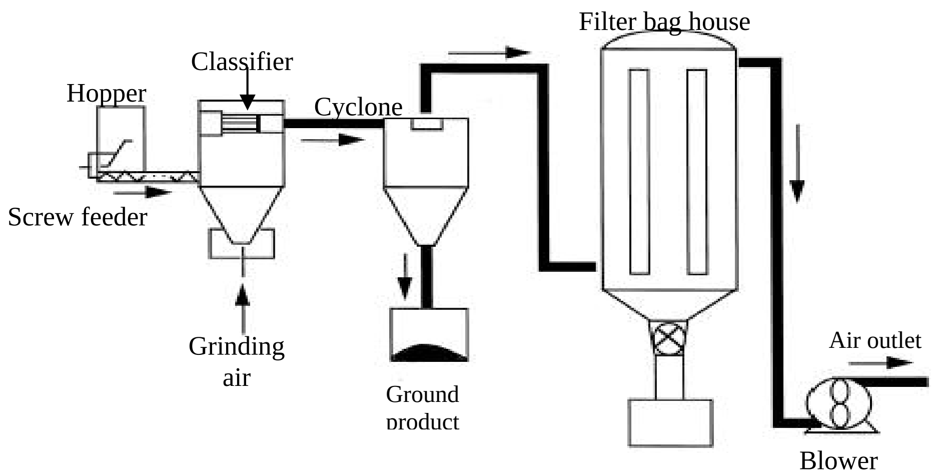

Figure 2.1: Schematic diagram of opposed fluidized bed jet mill (Berthiaux and Dodds,1999)

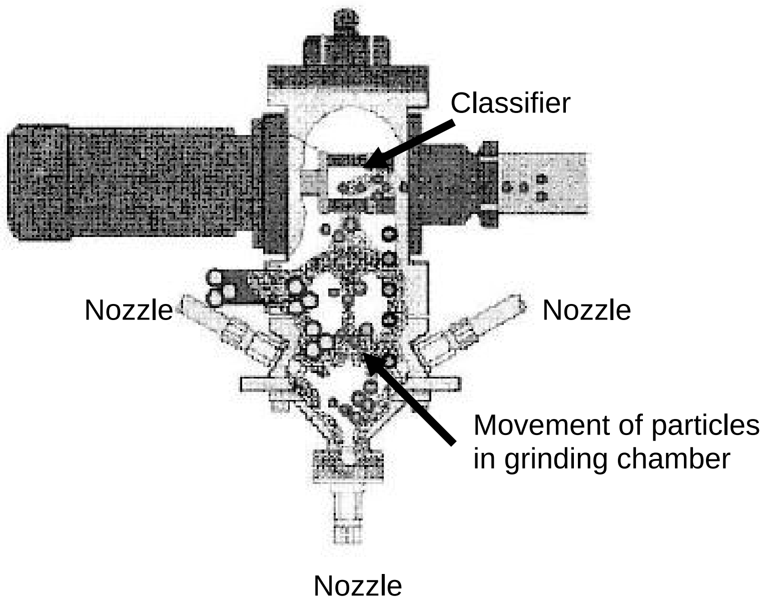

Figure 2.2: Grinding and classifying chamber of opposed fluidized jet mill (Berthiaux and Dodds,1999)

The feed for jet mill will be pre-milled and it will produce a mean particle size of less than 10μm depending on the design and operational parameters of the jet mill. The grinding fluid in the jet mill is usually air up to a pressure of 2 bars or steam up to 6 bars (Zhao and Schurr, 2002). If volatile material is being milled, the grinding fluid can be inert compressed gas such as nitrogen or carbon dioxide. Most industrial minerals are insensitive to temperature and thus the optimum size reduction can be achieved using high temperatures and high pressure super heated steam. This is typically in the range of 3 bars – 6 bars and 315^{o}C – 370^{o}C. However, with some minerals such as iron oxide based minerals pigments, the high temperature will affect the colour tint of the pigment. Typical grinding data of the jet mill on some minerals are shown in Table 2.2.

Table 2.2: Typical grinding data for jet mill (Russell, 1989)

Minerals	Mill diameter (cm)	Grinding fluid	Feed rate (kg/h)	d_{50} (μm)
Al_2O_3	20.3	Air	6.8	3
TiO_2	76.2	Steam	1020	<1
TiO_2	106.7	Steam	1820	<1
MgO	20.3	Air	6.8	5

Besides size reduction, excessive mechanochemical effect was observed in the ground particles due to the high energy intensity imposed on the particles and types of breakage mechanism that take place in the grinding chamber (Choi *et al.,*2004; Juhasz and Opoczky, 1990). The jet mill needs to be optimized due to the stringent demand of fine particles which are not only focus on the particle size distribution but also in terms of its crystallinity depending on its application. The inside mill condition such as the amount of hold up and the pressure inside the grinding chamber plays an important role especially for the opposed fluidized bed jet mill with internal classifier.

Nakach *et al.* (2004) disclosed the disadvantages of opposed fluidized bed jet mill such as poor particle size and its distribution, low capacity, expensive, mechanically complex and requires regular maintenance. The product quality from opposed fluidized bed jet mill was very much dependent on its characteristics, operational parameters and inside mill condition. An extensive and detail study of these factors is essential to obtain a good quality product which suits its application. The current stringent specification demand on fine particles which is used as mineral fillers has urged research focusing on crystallinity besides its particle size and distribution. Table 2.3 shows the application of jet mill in various sectors.

Table 2.3: Application of jet mill in various sectors (Russell, 1989)

Sector	Ground Product
Agrochemicals	Carbendezim, deltamethrine, fungicide, germicide, herbicide, sulphur.
Chemicals	Adipic acids, barium titanate, calcium chloride, catalyst, chrome oxide.
Ceramic	Aluminium hydrates, ferrites, glass, silicon carbide, zirconium oxide
Metals	Copper, molybdenum disulphide, noble metals.
Minerals	Bauxite, calcite, graphite, gypsum, mica, talc, tantalum ore.
Paints	Carbon black, fluorescent pigment, printing ink.
Pharmaceutical	Albendazole, antibiotics, aspirin. Bulk drug, cosmetics, omeprezole, oxfendazole.
Plastics	ABS resins, PVC stabilizers, phenolics, PTFE.
Others	Asbestos, chocolate, food colours, fuller earth, precipitated silica, wolframite ore.

2.4 Parameters affecting fine grinding process in jet mill

The design and operational parameters of fine grinding process in jet mill affects the ground product in terms of its product fineness, particle size distribution and the intensity of mechanochemical effect. The product fineness and its particle size distribution are affected by feed rate, grinding air flow rate, height of classification tube, angle of nozzles, types of grinding fluids, amount of holdup, grinding pressure and classifier frequency (Tuunila and Nystrom, 1998; Kolacz, 2004; Zhao and Schurr, 2002; Choi *et al.*, 2004; Gommeren *et al.*, 2000; Nakach *et al.*, 2004; Godet-Morand *et al.*, 2002). Juhacz and Opoczky (1990) reported the increased in the reactivity of the product ground in jet mill due to mechanochemical effect but the operating condition was not mentioned. Boldyrev *et al.* (1996) mentioned that mechanochemical effect was directly related to the operating condition of the grinding mill as the rate of stress and number of impulses on the particles would be determined by the operating parameters of the grinding mills.

The parameters which affect the product fineness in a jet mill can be classified as the geometrical parameters which is concerned with the design of the mill and operational condition. Table 2.4 shows the parameters that influence grindability in a jet mill (Midoux *et al.*, 1999).

Table 2.4: Geometrical parameter and operational condition of a jet mill (Midoux *et al.*, 1999; Benz *et al.*, 1996; Nykamp *et al.*, 2002; Nakach et. al., 2004)

Geometrical parameter	Operational condition
Diameter of grinding chamber	Solid feed rate
Shape of nozzles	Grinding pressure
Number of nozzles	Classifier speed
Angle of nozzles	Types of grinding fluids
Distance between the nozzles	Grinding aids
	Type of materials to be grind

2.4.1 Particle Breakage Mechanism in Jet mill

The breakage mechanism of particles in grinding mills is very much dependent on its design and operating parameters. In jet mill, the main breakage mechanism is destructive breakage due to impact and abrasion due to attrition between particles (Berthiaux and Dodds, 1999; Frances et. al., 2001). Figures 2.3 and 2.4 show the breakage mechanism of hydragillite and gibbsite respectively which exhibit the same particle breakage mechanism. This phenomenon indicates that the breakage mechanism of particles is influenced by the types of grinding mechanism. High grinding pressure (therefore, a higher jet velocity and greater grinding energy) leads to destructive breakage, therefore the product will be less block-shape and platelets but exhibiting sharp edges. If lower grinding pressure is used, abrasion grinding mechanism will be more dominant resulting in chipping of the particle edges.

(Berthiaux and Dodds, 1999; Tasirin and Geldart, 1999; Mebtoul *et al.*, 1996). Abrasion breakage mechanism will produce more rounded particles with less sharp edges.

Tasirin and Geldart (1999) found that coarse particles broke due to destructive breakage rather than abrasion and the probability of coarser particles chosen for breakage was higher than smaller particles. In general, smaller particles were more difficult to break than larger ones because smaller particles contained fewer faults, flaws or discontinuities.

Abrasion breakage mechanism in jet mill is dependent on the concentration or amount of particles in the grinding chamber. The increase in the amount of fines and creation of new surface area when the amount of holdup increases, is the characteristics of abrasion breakage mechanism taking place in jet mill (Mebtoul *et al.*, 1996). Abrasion breakage mechanism is characterized by formation of fines without noticeable change in mean diameter of particles.

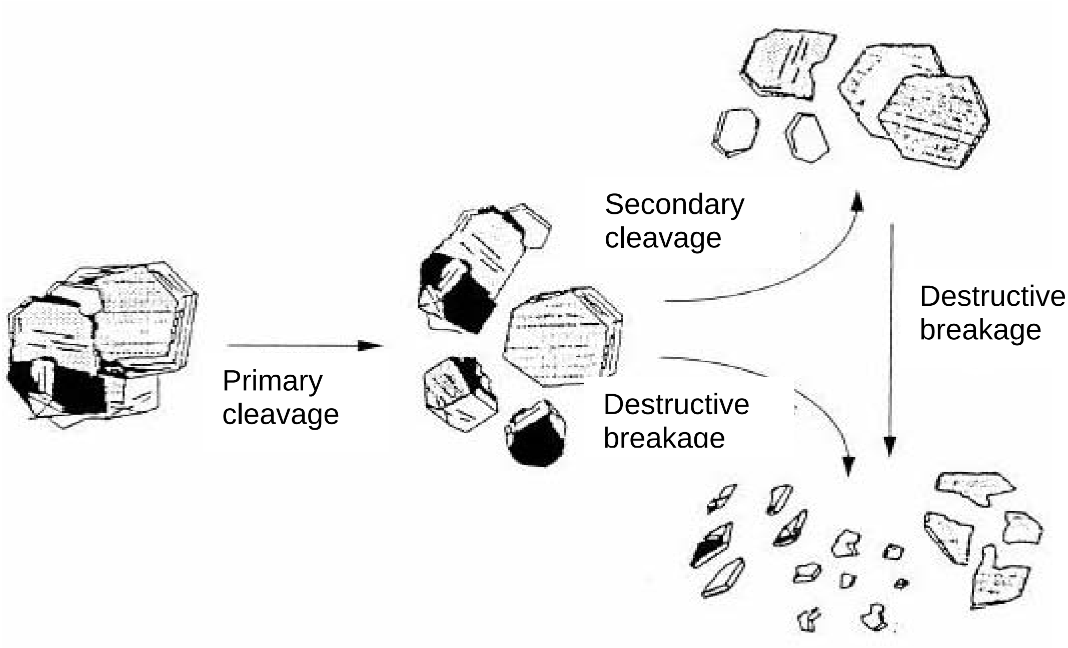

Figure 2.3: Schematic representation of breakage mechanism of hydrargillite particles in jet mill (Berthiaux and Dodds, 1999)

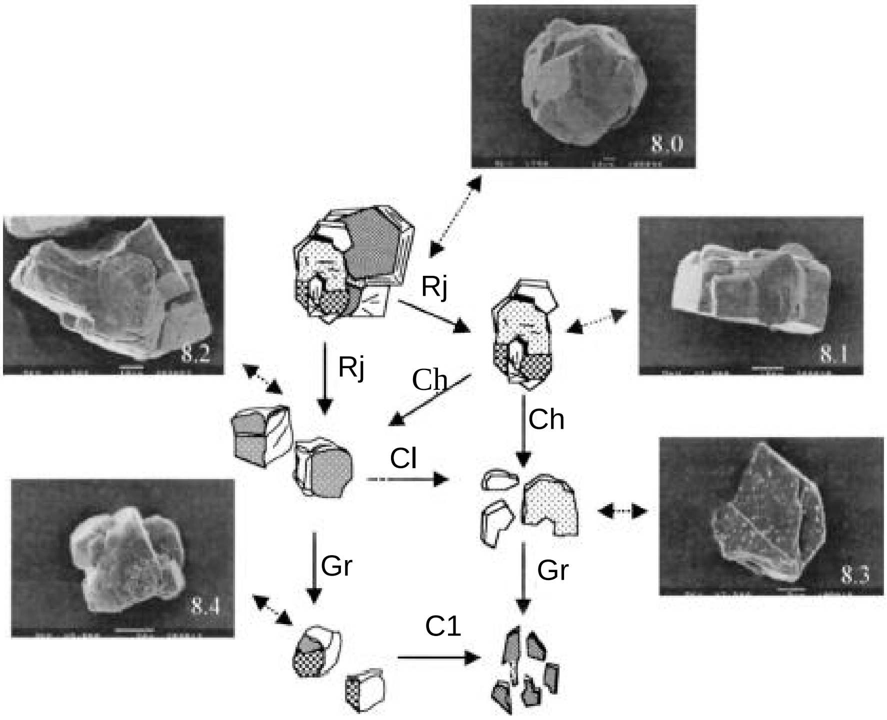

Figure 2.4: Fragmentation scheme for gibbsite ground in jet mill. Rj – rupture of joints, Ch – chipping, Cl – cleavage, Gr – ultimate grinding (Frances *et al.*, 2001).

Quantitative morphology analysis allows one to better evaluate the fragmentation processes (Lecoq *et al.*, 1999). There are several methods of particle quantification. Pons *et al.* (1999) classified three different levels of shape descriptors which are macroscopic descriptor, mesoscopic descriptors and pseudo-3D shape. The macroscopic descriptors are calculated from size measurements made on the particle silhouette.

The basic macro-shape descriptors are equivalent circular diameter, elongation and circularity. Equivalent circular diameter is the diameter of a disc having the same area as the projected silhouette. The calculation for equivalent circular diameter is shown in Equation 2.1.

$$D_{eq} = 2\sqrt{\frac{A}{\pi}}$$ **Equation 2.1**

The circularity (C) is a measure of both rugosity of the silhouette contour and its elongation as shown in Equation 2.2. Circularity is equal to 1 for a disk and increases when the silhouette departs from this reference shape, either because it gets elongated or its roughness increases. Circularity of a square is 1.27 (Marrot *et al.*, 2000) It should be used in conjunction with another shape factor such as elongation (Frances *et al.*, 2001).

$$C = \frac{P^2}{4\pi A}$$ **Equation 2.2**

The choice of the number of particles chosen for shape quantification is a critical issue as it will greatly influence the final remarks regarding shape (Faria *et al.*, 2003). Large number of particle need to be characterized to gain statistical validity and this process is very time consuming especially with sample preparation. This process will be more time consuming if the image capturing process is done in scanning electron microscope (Belaroui *et al.*, 2002). Therefore it is necessary to determine the number of particles to be examined to obtain statistically valid results. The effect of sample size on the morphological characteristics can be assessed by considering the changes of the descriptors average values and standard deviation versus number of particles. The average value of the descriptor is very essential to avoid the possibilities of shape orientation effect of the descriptors value as particle in the samples contains variety of values (Molina-Boisseau *et al.*, 2002). Belaroui *et al.*, (2002) determine the changes of the descriptors average values and standard deviation versus number of particles as shown in Figure 2.5. Belaroui *et al.* (2002) finally considered that 80 particles were sufficient enough to assess the morphology based on scanning electron microscopy photomicrograph.

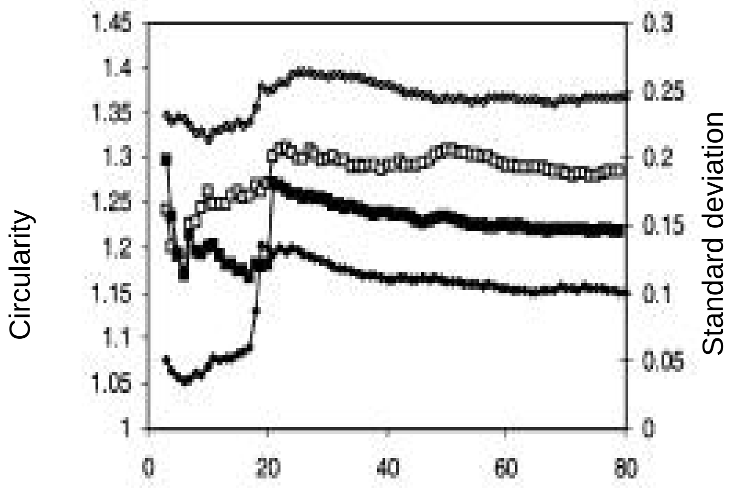

Figure 2.5: Variation of the circularity average values and the standard deviation (Belaroui *et al.*, 2002).

2.4.2 Jet mill design parameters

The jet mill design parameters are the angle of nozzles, types of nozzles, distance between the nozzles grinding chamber diameter and grinding chamber shape.

2.4.2.1 Effect of angle of nozzles

The research work on the angle of nozzles is limited. Only Tuunila and Nystrom (1998) and Midoux *et al.* (1999) reported the test work on angle of nozzles. Tuunila and Nystrom (1998) varied angle of nozzles ant three levels which were 23^0, 33^0 and 43^0. Tuunila and Nystrom (1998) reported that the angle of nozzles was less significant effect on the product fineness for two different types of gypsum but the maximum breakage was obtained at the largest angle which was 43^o as shown in Table 2.5.

Midoux *et al.* (1999) used three types of grinding mills with two different nozzle angles which were 63^o and 67^o. The nozzle angle depended on the size of the grinding chamber and had an effect on grinding ratio of the product (Midoux *et al.*, 1999). Furthermore, this angle affects the penetration of nozzle jets in the gas stream and it