Study on Wet Method Mechanism
for Producing Tronoh's Silica Sand Fine Particles

by

IZATUL NADIA BINTI ABU SAMAH

Dissertation submitted in partial fulfilment of
the requirements for the
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Universiti Teknologi PETRONAS
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CERTIFICATION OF APPROVAL

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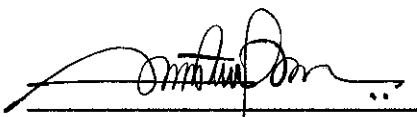
Approved by,



ASSOC. PROF. DR OTHMAN BIN MAMAT

UNIVERSITI TEKNOLOGI PETRONAS

TRONOH, PERAK

MAY 2011

CERTIFICATION OF ORIGINALITY

This is to certify that I am responsible for the work submitted in this project, that the original work is my own except as specified in the references and acknowledgements, and that the original work contained herein have not been undertaken or done by unspecified sources or persons.

IZATUL NADIA BINTI ABU SAMAH

ABSTRACT

There are abundant of silica sand found at Tronoh around the area of Universiti Teknologi PETRONAS, yet there is less effort for commercializing these silica sand in both technical and research application. The need to produce silica sand fine particles in various processes has increased tremendously. Thus, the objective of this project is producing and characterizing Tronoh's silica sand fine particles from wet method by ball milling using particle size analyser (Mastersizer), Scanning Electron Microscope (SEM), Field Emission Scanning Electron Microscope (FESEM), X-Ray Diffraction (XRD), X-Ray Fluorescent (XRF) and Energy Dispersive X-ray (EDX). The scope of study of this project is to produce Tronoh's silica sand fine particles using wet method by ball milling while characterizing the properties of materials formed using characterization equipment. It is based on both research and laboratory experiments, in which main equipment involved such as ball mill machine, sieve shaker and oven. The methodology of this project commences when silica sand is being taken at the entrance of Universiti Teknologi PETRONAS. The silica sand then being meshed with different of mesh sizes using sieve shaker until getting silica sand size <600um. The grinding process begins by mixing silica sand, water and grinding media to form slurry and grind it using ball mill. After that, silica sand is dried in an oven to remove water and moisture. The process of sieving, grinding and drying continue about 6 hours alternately until getting silica sand fine particles. From all these characterization equipment, the particle size distribution, particles morphology, crystalline structure, crystalline size, chemical composition and element mapping of silica sand fine particles can be determined. The results show that, as grinding duration increased, the particles size distribution of silica sand decreased, morphology structure changed, crystalline structure remained same with decreasing of crystalline size while composition of SiO_2 increased and both weight and atomic percent of Si element decreased. These characteristics of silica sand fine particles are very imperative both for commercialization and advance research. I believe that my final year project will be beneficial to both UTP and industry.

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"Study on Wet Method Mechanism for Producing Tronoh's Silica Sand Fine Particles"

To all the Graduate Assistance (GA) as well as all technicians, I would like to extend a heartfelt thank you for the guidance and effort in helping me with all their technical expertise and facilities.

Special thanks to my family, housemates and fellow friends for keep me calm; help me when pressure comes and also in giving morale support.

Last but not least, I would like to thank all individuals who have contributed in one way or another in initiating this project.

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TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

CHAPTER 1
INTRODUCTION

1.1 Project Background

This Final Year Project (FYP) is focusing on development of Tronoh silica sand fine particles using wet milling mechanism. Silica is in abundant found in crust of earth. In fact, Tronoh, Perak has abundance of silica sand. Nevertheless, people take it for granted and less effort for commercializing this silica sand. Therefore, in this project, ball milling will be used to produce silica sand fine particles using wet milling mechanism.

The most popular top-down solid material is the grinding method. Ball mill is an efficient tool for grinding many materials into fine powder. The grinding media, zirconium ceramic beads are used in ball mill to impart the forces required for size reduction. Particle sizes from just submicron to some 100 microns can be achieved using either dry or wet method process. However, for smaller particles or fine particles, wet method is used. The options to choose whether dry or wet method influenced by several factors. The grinding process can be determined by the condition of the raw material and the downstream use as powder or slurry. Other subjects like product fineness, dispersion, hazards or chemical reactivity have to be considered [1]. In this project, the mechanism that will be used is wet method. Water is used as the wetting agent.

The silica sand particles obtained will be characterized using advanced technology equipment such as particle size analyzer (Mastersizer) for particle size distribution, Scanning Electron Microscope (SEM) and Field Emission Scanning Electron Microscope (FESEM) for particles morphology, X-Ray Diffraction (XRD) for crystalline structure and size, X-Ray Fluorescent (XRF) for chemical composition and Energy Dispersive X-ray (EDX) for element mapping.

1.2 Problem Statement

1.2.1 Problem Identification

There are abundant of silica sand found at Tronoh around the area of Universiti Teknologi PETRONAS, yet there is less effort for this silica sand in technical and research application.

1.2.2 Significant of the Project

The experimental project is significant in producing Tronoh's silica sand fine particles using a low speed ball mill having speed up to 90rpm by using zirconium ceramic beads as grinding media. The study gives deep understanding on wet method and characterization of silica sand fine particles produced.

1.3 Objective and Scope of Study

1.3.1 Objective of project

Producing and characterizing Tronoh's silica sand fine particles from wet method by ball milling using particle size analyser (Mastersizer), Scanning Electron Microscope (SEM), Field Emission Scanning Electron Microscope (FESEM), X-Ray Diffraction (XRD), X-Ray Fluorescent (XRF) and Energy Dispersive X-ray (EDX).

1.3.2 Scope of study

In this project, the scope of study is to produce Tronoh's silica sand fine particles using wet method while characterizing the properties of materials formed using particle size analyser (Mastersizer), Scanning Electron Microscope (SEM), Field Emission Scanning Electron Microscope (FESEM), X-Ray Diffraction (XRD), X-Ray Fluorescent (XRF) and Energy Dispersive X-ray (EDX). It is based on both research and laboratory experiments, in which main equipment involved such as ball mill machine, sieve shaker and oven.

1.4 The Relevancy of the project

Since there are abundant of silica sand found in Tronoh, thus it is very meaningful to use the silica sand and process them using ball mill by wet method to produce silica sand fine particles. As the particles size is reduced, it will increase in surface area and surface to bulk atom ratio. The increase in number of surface atoms changes the physical and chemical properties of the materials. Thus, some of the important properties such as particle size distribution, particles morphology, crystalline structure and purity can be determined by using particle size analyser (Mastersizer), Scanning Electron Microscope (SEM) and Field Emission Scanning Electron Microscope (FESEM), X-Ray Diffraction (XRD), X-Ray Fluorescent (XRF) and Energy Dispersive X-ray (EDX). Since all these equipment as well as the raw material are available at Universiti Teknologi PETRONAS, thus this project is relevant to be carried out.

1.5 Feasibility of the Project within the Scope and Time Frame

It is an obligatory for mechanical engineering students to complete final year project within 2 semesters. The project commences with research work in first semester (FYP 1) following by experimental work in second semester (FYP 2). It will be assumed that the project is feasible within the scope and time frame regardless of no issues with regard to equipment function and material availability.

CHAPTER 2
LITERATURE REVIEW

2.1 Ceramic

Ceramic cover large range of materials whether traditional ceramics or advanced ceramics. Traditional ceramics relates the development of ceramic since the time of earlier civilizations such as pottery, clay products and clay-based refractory. Nowadays, industry has shown more interest in advanced ceramic include for electrical, magnetic, electronic and optical application [1].

Normally, ceramic are crystalline ceramics or also called polycrystalline [20]. They are made up a large number of small crystals or grains separated from one another by grain boundaries.

There are two broad types of ceramic raw materials that require milling. They are classified as lumpy and powdered ceramics. Lumps result from mining, fusion, and sintering. These are pre-milled by supplier and are available in various screen sizes. Depending o requirements, these require further milling in the lab. Mining materials include talc, shale, bauxite and quartz. Fused materials include fused alumina, magnesia, mullite and zirconia [24].

Meanwhile, they are two structure type of ceramic which are at the atomic scale and at the large scale (microstructure) [20]. At atomic scale, the type of bonding and the crystal structure (for crystalline ceramic) or amorphous structure (for glassy) play an important definition. Whereas, at larger scale or microstructure refers to the nature, quantity and distribution of the structural elements or phases in the ceramics.

2.2 Silica Sand

Large quantity of the earth covers by silica sand and are the source from which most silica is made. Different ultrafine particles of silica with different sizes prepared mainly from the corresponding precursors.

Silica sand can be fabricated into two main methods which are gas-phase reactions method (drying method) and liquid precursor methods (wet method) [20]. The particles sizes in both the gas phase and the liquid precursor method are quite different in that the former produces silica with a smaller particle size than that produced by the latter method [2].

Table 2.1 shows characteristic index of silica sand where the particle sizes are less than 40nm. Silica sand has been produced in large amount to be industrialized. Currently, silica sand has been produced in field of coating, engineering plastics, automobile tires and electronic components. Silica sand has high specific surface area and hydrogen content [2].

Table 2.1: Characteristic index of silica

Parameters	Index
Specific surface area (m^2g^{-1})	640 ± 50
Particle size (nm)	$(10–30) \pm 5$
Silica content (%)	> 99.9
Stirring density (gm^{-3})	< 0.15
Impurities (%)	$Cl < 0.02$, metal < 0.001
Content of surface hydrogen (%)	(30–40)

"Background on Polymer-Layered Silicate and Silica Nanocomposites: Characteristic Index for Silica"[1].

2.3 Fine Particles

In the materials world, especially ceramics, the trend is always to prepare finer powder for ultimate processing and better and useful properties for various applications. The fineness can reach up to molecular level by special processing techniques. More is the fineness; more is the surface area, which increases the 'reactivity' of the material. Fine particles characterization or powder characterization depends strongly on the method used to synthesize them thus influencing the subsequent processing of ceramic.

The powder characteristics of greatest interest are the size, size distribution, shape, degree of agglomeration, chemical composition and purity. Table 2.2 illustrates the desirable powder characteristics for advanced ceramics. Usually, a powder with a wide distribution of particle sizes (sometimes referred to polydisperse powder) generally leads to higher packing density [20].

A common problem is that if the particles are not packed homogeneously to give uniformly distributed fine pores, densification in the later stages can be slow. The larger grains coarsen rapidly at the expense of the smaller grains, making the attainment of high density with controlled grain size impossible. Homogeneous packing of a narrow size distribution powder (nearly monodisperse powder) generally allows better control of the microstructure. A spherical or equiaxial particle shape is beneficial for controlling the homogeneity of the packing [20].

Table 2.2: Desirable Powder Characteristics for Advanced Ceramics

Powder Characteristic	Desired Property
Particle size	Fine (< ~1 μm)
Particle size distribution	Narrow or monodisperse
Particle shape	Spherical or equiaxial
State of agglomeration	No agglomeration or soft agglomerates
Chemical composition	High purity
Phase composition	Single phase

"Synthesis Powder: Desirable Powder Characteristics for Advanced Ceramics" [20].

2.3.1 Problems of Preparation Fine Particles

The preparation of fine-crystalline particles is very difficult problem. The properly distributed fine crystalline particles within the very narrow range by grinding necessitate a proper understanding of the process of grinding.

At initial stage, large cracks in original particles propagate, generating smaller particles with very fine cracks. However, the probability of particles having cracks progressively decreases with grinding leading to the generation of newer stronger particles [24]. Hence continuously higher and higher fracture stress is required to grind the particles. During the grinding, it becomes progressively difficult to obtain further reduction in particle size on fine crystalline region.

In fine crystalline region, the fine particles develop the tendency to agglomerate and the physical equilibrium between the aggregates and fragmentation is established, resulting in decrease in 'storing of stress energy'. Consequently, it results in an increased stress for initiating fracture and the generation of fine particles. Agglomerates lead to heterogeneous packing in turns lead to different region of the body shrink at different rates. This leads to serious problems such as development of large pores and crack-like voids. Agglomerates can be classified into two types which are soft agglomerates in which particles are held together by weak van der Waals forces and hard agglomerates in which the particles are chemically bonded together by storing bridges. The ideal situation is to avoid agglomeration. Nevertheless, in most cases this is impossible. Thus, soft agglomerates is more preferable compared to hard agglomerates as soft agglomerates can be broken down easily by mechanical methods (pressing or milling) or by dispersion in a liquid.

There are two principal factors which need to be controlled to obtain fine particles:-

1. Ratio of weight of the balls or grinding media to the total weight of the particles to be processed by the grinding route.
2. Semi-fluid/viscous characteristic of the grinding media containing fine particles.

2.4 Methods Producing Silica Sand Fine Particles

2.4.1 Wet Method Mechanism

One of the ways to produce fine particles silica sand is by using wet method mechanism. Wet method is the process of adding a liquid solution to powders. It is one of the most common ways to granulate. Wet method mechanism is done when silica sand is added with wetting agent to produce fine slurry. Wetting agent makes the particles of silica sand and ceramic beads stick together during the grinding process. As a result, grinding energy efficiency is improved and less time requires producing the fine particles.

Normally the typical wetting agent content is approximately 50-60%. The wet process had a number of advantages. Wet grinding of hard minerals is usually much more efficient than dry grinding. When slurry is dried, it forms a granular crumble that is ideal for subsequent heating. In the dry process, it is very difficult to keep the fine powder mix. Typical liquids used include water, ethanol and isopropanol either alone or in combination. The liquid solution can be either aqueous based or solvent based. Aqueous solutions have the advantage of being safer to deal with than solvents [3].

Normally, water will be used as wetting agent since it has been showed from other experiment to produce fine particle with short in time compare to other liquids. Water mixed into silica sand can form bonds between powder particles that are strong enough to lock them together. The process can be very simple or complex depending on the characteristics of the powders, the final particles obtained, and the equipment that is available. In the traditional wet method is when the wet mass is forced through a sieve to produce wet granules which are subsequently dried [3].

Wet ball mill has somewhat higher energy utilization (10-20%) than dry milling and the ability to produce a higher fraction of finer particles. On the other hand, wet milling suffers from increased wear of the grinding media, the need for drying the powder after milling and the possible contamination of the power by adsorbed molecules from liquids medium.

2.5 Equipment / Materials Used for Producing Silica Sand Fine Particles

2.5.1 Ball mill

The usual objective of reducing the size of material is to separate the mineral interest contained in the ore body from associated gangue minerals. Crushing only does not generally liberate silica sand into fine particle. There needs further size reduction. This can be achieved by grinding the silica sand in ball mill. Ball mill is a type of grinder which is a device used in grinding materials like ores, chemicals and ceramic raw materials. Different materials are used as grinding media to assist grinding, including ceramic balls, flint pebbles and stainless steel balls [11]. The grinding media, zirconium ceramic ball is used to impart the forces required for size reduction. As a result of combined action of repeated impact and abrasion over time, size reduction will take place. The process of ball mill commences when the material needs to be grinded is put into the ball and the grinding ball is then rotated over and over [12].

The rate of grinding depends on several factors such as mill parameters, the properties of the grinding medium and the properties of the particles to be ground [20]. Generally, ball mills that run at low speeds contain large balls because most of the mechanical energy supplied to the particle is in the form of potential energy. Meanwhile, ball mills that run at high speeds contain small balls because most of the energy supplied to the particles in the form of kinetic energy.

The rate of grinding also depends on the particle size. The rate decreases with decreasing particle size as the particle become fairly fine, it became more and more difficult to achieve further reduction in size. A practical grinding limit depends on several factors. First, the increased tendency for the particles to agglomerate with decreasing particle size means that a physical equilibrium is set up between the agglomeration and comminution processes. Second, the probability for the occurrence of a comminution event decreases with decreasing particle size [20]. The longer the ball mill runs, the finer the powder will be. Ultimate particle size depends entirely on how hard the grinding material, and the time how long the ball mill runs.

There are several advantageous of using ball mill which are:-

- Suitable for grinding material with high hardness
- No pollution for the powder with ceramic ball
- Stable performance
- Easy installation
- The capacity and fineness can be adjusted by adjusting the diameter of the ball.

A disadvantage of ball milling is that wear of the grinding medium can be fairly high [20]. For advanced ceramics, the presence of impurities in the powder is a serious problem. The best solution is to use balls with same composition as the powder itself. However, this is only possible in very few cases.

Another solution is to use grinding media that is chemically inert at the sintering temperature of the ball (ZrO_2) or can be removed from the powder by washing (steel balls). A common problem is the use of porcelain balls or low purity Al_2O_3 balls that wear easily and introduce a fair amount of SiO_2 into the powder.

2.5.2 Sieve Shaker

The particle size distribution (PSD) of a powder, or granular material, or particles dispersed in fluid, is a list of values or a mathematical function that defines the relative amounts of particles present, sorted according to size [17]. The way PSD is expressed is usually defined by the method by which it is determined.

The most easily understood method of determination is sieve analysis, where powder is separated on sieves of different sizes [17]. Sieve analysis is accomplished by passing a known weight of sample material successively through finer sieves and weighing the amount collected on each sieve to determine the percentage weight in each size fraction. Sieving is carried out with wet or dry materials and the sieves are usually agitated to expose all particles to the opening [3].

The process of sieving can be characterized into two which are:-

1. Elimination of smaller particles than the screen apertures
2. Separation of the so-called "near-size" particles

Both rewires sieve to be manipulated in that all particles have chance to pass through the apertures.

Machine sieving is almost universally used as hand sieving is long and tedious. Sieves can be arranged in the range of diameters, depending on the particle size and mass of material to be sieved [3]. A common diameter for laboratory sieves is 200mm.

The sieves chosen for the test are arranged in a stack, or nest with the coarsest sieve on the top and finest at the bottom. A tight-fitting pan or receiver is placed below the bottom sieve to receive the final undersize and lid is placed on top of the coarsest sieve to prevent escape of the sample. The material to be tested is placed in the coarsest sieve and the nest is then placed in sieve shaker which vibrates the material in the vertical plane and some on horizontal plane.

During the shaking, the undersize material falls through successive sieves until it is retained on the sieve having apertures which are slightly smaller than the diameter of the particles. In this way the sample is separated into size fractions. The nest is then taken out and the amount of retaining material on each sieve is weighed. Most of the near-mesh particles which block the openings can be removed by inverting the sieve and tapping the frame gently. At the completion of the test the sieves together with the retained oversize material, are dried at the suitable low temperature and weighed.

The advantageous of sieve shaker are as follows:-

- The material moves evenly over the entire sieving surface resulting in short sieving times and accurate separation
- Accurate results from test to test

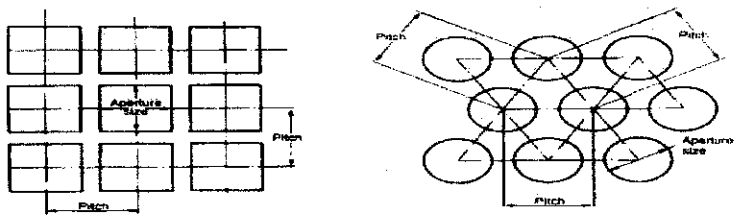


Figure 2.1: Arrangement of square ad round holes in perforated plate sieves [3]

2.5.3 Zirconia Grinding Media

The term, "grinding media" refers to the items that are used to do the grinding inside of the grinding ball. There are different types of grinding media because of the different types of materials to be ground. Composition of media includes porcelain, high alumina, pure alumina, TZP, MgO stabilized zirconia, silicon nitride, silicon carbide, steel, modified fused zircon and variety of mineral products such as flint, agate or material that is ground by itself [24]. Table 2.3 shows some of the commercially available grinding media for ball milling

Normally, type of grinding media chooses is Zirconia. Zirconia grinding media is the most durable and efficient media for ball milling of ceramic materials. Zirconia grinding media provides a virtually contamination-free ball milling solution for a variety of industries. As mass is proportional to the density, the grinding medium should consist of materials with as high density as possible.

The size of grinding media also plays an important role. Small grinding media are generally better than larger ones. The number of balls increases inversely as the cube of the radius. Thus, the rate of grinding depends on the number of contact points between the balls and the powder. The number of contact in turn depends on the surface area of the balls. Hence, the rate of grinding will increase inversely as the radius of the balls. However, the balls cannot be too small as they must impart sufficient mechanical energy to the particles to cause fracture.

The advantageous of Zirconia as the grinding media are:-

- High density, high tenacity, impact-resistance and low wear.
- Smooth surface
- Ultra low wear loss as a result of the transformation-toughening mechanism unique to the stabilized zirconia material system.
- Higher grinding efficiency and reduced grinding time due to zirconia's higher density (compared with alumina and glass).
- Most durable grinding material, reducing operational cost in the long run. Wear rate substantially lower than Al_2O_3, CeO_2-ZrO_2, and MgO-ZrO_2 grinding media.
- Suitable for high velocity operations and wet grinding operations.

Criteria in Selecting Size of Zirconia Grinding Media:-

- Larger-sized media are more efficient in breaking up large particles.
- Smaller media are more efficient in milling smaller particles because of higher effective contact area.
- Larger media sizes are favored in many conventional ball mills. Smaller sizes are more appropriate for attritors. For example, 5mm media are used in many attrition mill applications, whereas 10-15 mm may be a good start for a lab ball mill. For small-media mills, media sizes are typically smaller than 2 mm.

Table 2.3: Commercially Available Grinding Media for Ball Milling

Commercially Available Grinding Media for Ball Milling

Grinding Media	Density (g/cm3)
Porcelain	2.3
Silicon nitride	3.1
Silicon carbide	3.1
Alumina	
Lower than 95% purity	3.4–3.6
Greater than 99% purity	3.9
Zirconia	
MgO stabilized	5.5
High-purity Y_2O_3-stabilized	6.0
Steel	7.7
Tungsten carbide	14.5

"Synthesis Powder: Commercially Available Grinding Media for Ball Milling" [20].

2.6 Crystalline and Amorphous phases

The solid phase in engineering materials can be either crystalline or amorphous. Amorphous phase forms from rapid cooling from the liquid phase, condensation from gaseous phase or result of chemical reaction. The term crystalline comes when the local atomic packing results in long-range order whereas amorphous term comes when the local atomic packing results in short range atomic order and has no long-range consequences [15].

2.7 Journals / Papers

Some of the journals related to this project have been studied and summarized.

First and foremost, the paper studied from T. Ahmed & O. Mamat [6] on the production of silica sand nanoparticles by using low speed ball mill. The result produced by "size distribution by intensity" generated from Zetasizer Nano Analyser shows that the average silica sand particle size produced was 78.82nm. Meanwhile, results from X-Ray Diffraction shows that the lattice diameter of silica sand particles is 60.007nm. From results of Field Emmision Scanning Electron Microscope (FESEM), after 6 hours of grinding, it shows that agglomerations of the nanoparticles were clearly observed which the average size of silica sand particles of 361nm and 162nm. EDS analysis on the silica sand revealed the presence of high Si content with little amount of Al.

The second paper is from M. Santos [4]. The research is regarding high energy milling behaviour of alpha silicon carbide. It is said that alpha silicon carbide powder was comminuting in planetary mill during the periods of time of ½, 2, 4 or 6h. The rotation speed was 300rpm, the milling media used was isopropilic alcohol and the grinding bodies were spheres of zirconia established with ceria. It was observed a great reduction in the particle size, from micrometric to submicrometric size and even nanometric particles. The XRD result shows the presence of small broad peaks.

The paper from K.Y. Rhee, H.K. Cho & J.S. Hong [5] which the tittle of an investigation on the application of cryogenic ball milling to ibuprofen particle and it characteristics shows the results of reduction of ibuprofen particles size about 1/20 of its initial size. However, after six hours of ball milling, the particle size showed negligible changes. No chemical changes during process and changes in intensity and width of XRD pattern shows result of reduction in particle sizes.

Furthermore, paper from K. Akcay, A. Sirkecioglu, M. Tather, O. Savasci & A. Senatalar, regarding wet ball milling of zeolite HY [21]. The result from this paper shows that after 2 hours of wet ball milling, the medians of the particle size distribution curves by volume and by number could be reduced from about 6 and 2 μm to about 1 μm and 70 nm, respectively. It also said that even after 14 h of grinding, the characteristic X-ray diffraction peaks of zeolite HY were still observable. It was concluded that the crystalline structure could be partly retained even after quite long milling times.

Moreover, paper from N. Kotake, M. Kuboki, S. Kiya & Y. Kanda [22] shows that the median particle sizes of product obtained for the grinding limit in wet and dry conditions were around 0.5–0.6 μm and 2 μm, respectively. The width of the particle size distribution in wet grinding decreased with decreasing median particle size of the ground product, and the size distribution in dry grinding became nearly constant.

Last but not least, the paper from C.C Koch [16] which the tittle of top-down synthesis of nanostructure material shows that only ball milling, high pressure torsion and accumulative roll bonding can have regular average grain sizes below 100nm – that is nanocrystalline material.

CHAPTER 3
METHODOLOGY

3. 1 Project Background

Proper techniques need to be designed and done to attain the objectives of this project. This project is regarding the study on wet method mechanism for producing Tronoh's Silica Fine Particles using ball mill. This project needs to be completed within 2 semesters in final year. The Gantt chart for both Final Year Project 1 and Final Year Project 2 are shown on Table 3.4 and Table 3.5.

3.1.1 Problem Identification & Literature Review

First and foremost, the problem that caused the project to be done need to be identified. Then, the specific task for the project commences with literature review of the project. Literature review need to be done by finding all the journals, conferences, proceedings or even books regarding the project.

3.1.2 Silica Sand Collection

The practical of the project commences by collecting an amount of silica sand outside Universiti Teknologi PETRONAS entrance at Tronoh, Perak. The silica sand was taken by digging two to three feet deep hole from the surface. This will ensure the silica sand taken is less mix up with contamination exists surrounding the place taken. Figure 3.1 show the silica sand taken.

Figure 3.1: Silica sand taken

3.1.3 Pre-experiment

The process proceeds with the pre-experiment. About 1kg of silica sand was measured using electronic weigh (>500g). The silica sand then being dried in the oven for one hour with 110°C temperature to remove its moistures. Figure 3.2 shows the silica sand being dried in an oven. Meanwhile, Figure 3.3 shows silica sand after drying which consists of both course and fine particles. Silica sand produced consists of both large and fine particles. Therefore, meshing need to be done to segregate silica sand at different size of meshing.

Figure 3.4 shows the arrangement of pans with different in sizes. The meshing is done by using Sieve Shaker model EFL 2000. Figure 3.5 shows the Sieve Shaker that being used to sieve silica sand. It is a must for the silica sand to be meshed in order to segregate silica sand between large and small size. Otherwise, time taken to grind the silica sand using ball milling will be long. The silica sand that have been dried will be sieved with different size of pans until getting size of less than 600 μm in duration of 10 minutes.

Figure 3.2: Silica sand being dried in an oven

Figure 3.3: Silica sand after drying consists of both course and fine particles

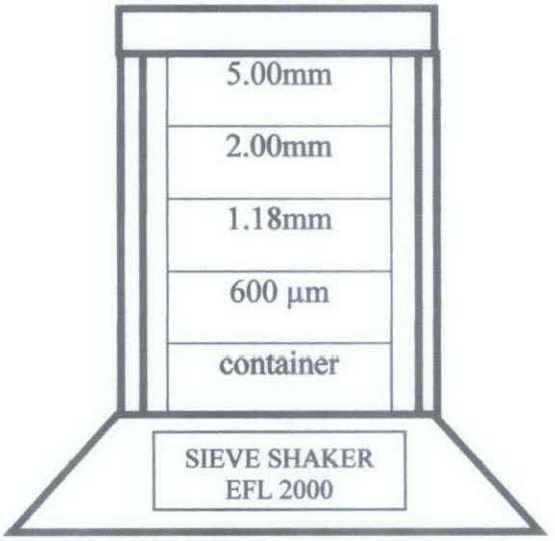


Figure 3.4: Arrangement of pans with different in sizes (pre-experiment)

Figure 3.5: Sieve Shaker EFL 2000

3.1.4 Ball milling

The technical experiment starts by grinding silica sand by wet method mechanism using ball milling. Ball milling machines used is US STONEWARE 764AVM Ball Mill. It is totally enclosed with DC drive motors permit solid-state control of roll speed. This machine has the speed range of between 20 and 300 RPM. The motor is powered with 1/4 horse power (HP) and furnished with standard plug. In wet method, water is used as a wetting agent. Meanwhile, the grinding media used is zirconium ceramic beads. The grinding media ratio towards weigh of silica sand used in this project is 10.0:1. 60ml of water, 100g of silica sand and 320 pieces of grinding media are mixed to form slurry inside a jar mill.

The parameters used for grinding silica sand are shown in Table 3.1. Figure 3.6 shows the mixture of water, silica sand and grinding media to form a slurry. The jar mill then is being put on the mill rack of ball mill machine. The ball mill that being used to grind the mixture is shown in Figure 3.7 It is important to ensure that the opening of the jar mill is fully closed as to avoid any leakage during the rotational process. Gasket is placed on the opener of jar mill to produce a perfect fit. Thus, during vibration caused by milling, the slurry inside the jar mill will not splash up to the gasket, so eliminating contaminations.

The grinding process has been divided into six different stages. Table 3.2 shows the stages taken to grind silica sand with different pans size. For each stage, silica sand is ground using ball mill for one hour duration with speed of 90rpm.

In order to produce silica sand fine particle, the ball milling process is repeated to five times until getting very fine silica sand. Each of the alternate stage, silica sand is meshed using Sieve Shaker at different pan sizes. However, Sieve Shaker model EFL 2000 is capable to sieve as small as 63 μm. Thus, in order to obtain very fine silica sand, the silica sand at last stage (5th stage) will be ground for 2 hours.

Table 3.1: Parameter used for grinding silica sand

Parameter	
Grinding media ratio towards silica sand weight	10.0 : 1
Zirconium beads (grinding media)	320 pieces
Silica sand weight	100 g
Water contain	60 ml

Figure 3.6: Water, silica sand and grinding media are mixed to form a slurry

Figure 3.7: US STONEWARE 764AVM Ball Mill and jar mill

Table 3.2: Stages taken to grind silica sand with different pans size

Stage	Grinding Duration (Hours)	Size of Pans (μm)
1st	1	<425
2nd	1	<212
3rd	1	<150
4th	1	<63
5th	2	-

3.2 Equipment for Characterization

The silica sands produced are being analysed using characterization equipment. The evaluation of the effects of grinding duration on particle size distribution and morphology evolution of silica sand were carried out by particle size analyzer (Mastersizer) and both Scanning Electron Microscope (SEM) and Field-Emission Scanning Electron Microscope (FESEM) respectively. The changes in crystalline structure and size of silica sand during grinding are studied by X-Ray Diffraction (XRD) patterns.

Meanwhile, the chemical compositions of silica sand are studied by using X-Ray Fluorescence (XRF) whereas elemental mapping of silica sand is determined using Energy Dispersive X-ray (EDX). All of these characterizations are done at 0 hours, 2 hours, 4 hours and 6 hours of grinding duration. Table 3.3 shows the equipment and characterizations used in this project.

Table 3.3: Equipment and characterizations used

Equipment	**Characterizations**
Sieve Shaker (Sieve Analysis)	mesh size of silica sand
Particle Size Analyzer (Mastersizer)	size distribution of particles
Scanning Electron Microscope (SEM)	morphology evolution at 1000x magnification
Field-Emission Scanning Electron Microscope (FESEM)	morphology evolution at 20Kx magnification
X-ray Diffraction (XRD)	crystalline structure and size
X-Ray Fluorescence (XRF)	chemical composition
Energy Dispersive X-ray (EDX)	elemental mapping

3.3 Flow Chart for Processing Silica Sand

Figure 3.8 shows the flow chart for processing silica sand.

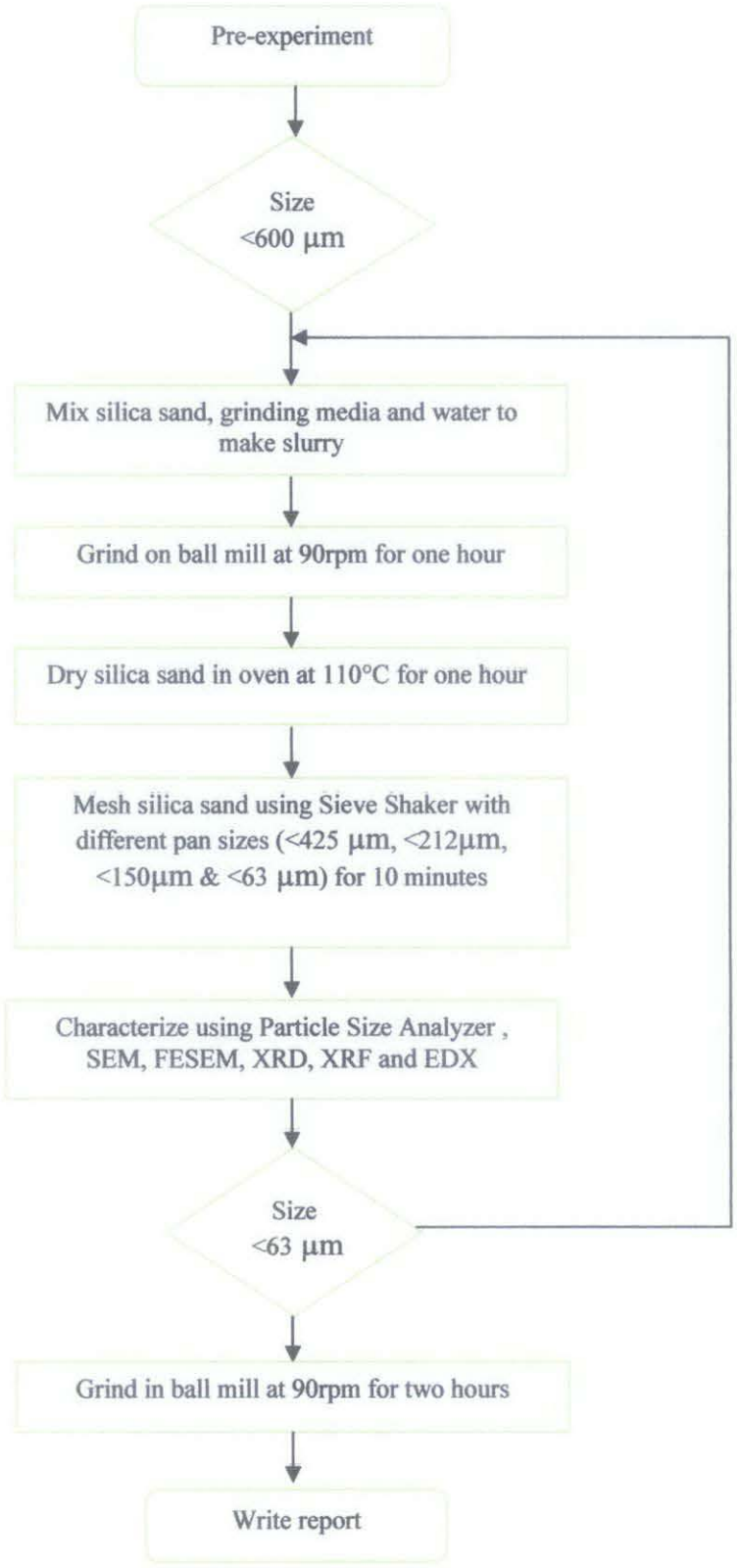


Figure 3.8: Flow chart for the processing of silica sand

3.4 Characterization

3.4.1 Particle Size Distribution

The particle size distribution of silica sand can be determined by using Mastersizer 2000 particle size analyser. The Mastersizer 2000 laser diffraction-based particle size analyzer has advanced particle size distribution measurement. It can measured materials size in range of 0.02 μm to 2000 μm.

The particle size may also vary over quite a wide range. It is not unusual for the particles of a suspension produced in a grinding operation, for example, to vary by a factor of 100 from the smallest to the largest size. To describe such situations we normally break the range up into a number of classes and try to find out how many particles are in each size range. This range is called the particle size distribution (PSD), and it can be represented in the form of a table or a histogram. Figure 3.9 shows Mastersizer 2000 particle size analyser.

Figure 3.9: Mastersizer 2000 particle size analyser

3.4.2 Morphology Evolution

The primary used of SEM is to study the surface topography of solid samples. SEM is used to observe microstructure when the grain size is too small for optical microscopes. SEM imaging is preferred in place of optical imaging because of the enhanced depth of field. Organic samples need to be covered with a layer of electrical conductive substances (e.g., metal or carbon) to avoid charging effects during imaging. Also, the energy of the electron beam has to be adjusted accordingly to avoid damaging the specimen [23]. Figure 3.10 shows Scanning Electron Microscope (SEM).

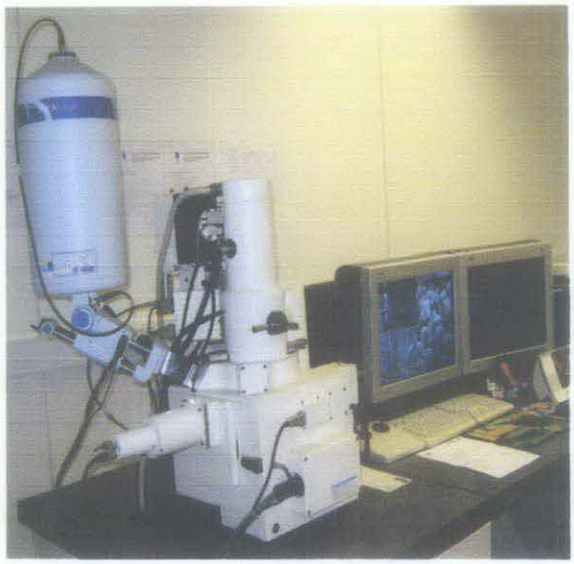


Figure 3.10: Scanning Electron Microscope (SEM)

Since most ceramics are electrically insulating, at higher magnification, surface charging occurs and blurs the image. Thus, Field-emission SEM (FESEM) instrument is used for materials characterization. The use of a high-brightness field-emission gun makes it possible to acquire nanometre-resolution surface images at low voltages (<5kV) [10]. Using FESEM analysis, the morphology of silica sand particles can be obtained with higher magnification. The images of the sample surface are revealed by scanning it with a high-beam of electrons [19]. Figure 3.11 shows Field-Emission Scanning Electron Microscope (FESEM).

Figure 3.11: Field-Emission Scanning Electron Microscope (FESEM)

3.4.3 Crystalline Structure and Size

X-ray diffraction (XRD) method is a method for characterizing the crystal structures and size. It is employed when a powder or polycrystalline specimen consisting of many fine and randomly oriented particles that are exposed to monochromatic x-radiation [14]. Whether the structure of silica sand is crystalline or amorphous, XRD can determine it. The XRD methods are based on the measurements of X-ray intensities scatters. Diffraction occurs not only in double split, but also in any periodic structures such as crystal lattices [7]. In X-ray measurements, the diffraction peak position, half-width and identification of peaks are the main parameters used to describe materials [13].

The X-Ray Diffraction (XRD) was recorded with D8 Advance, Brucker AXS Diffraction using Ni-filtered Cu Ka Radiation (λ=1.5406 Åm) operated at 9kv of the step scan size 0.020 2θ and counting time 10 seconds in the range of 2θ of 100 to 800. The XRD analysis identifies the values of full width half maxima (FWHM), d-spacing and crystalline size for the [1 1 0] plane as a high peak. Diffraction occurs not only in double split, but also in any periodic structures such as crystal lattices [18].

Changes in the XRD patterns reflect the evolution with time of the phases during milling of silica sand. The nature of the phases present and the general shape of the peaks depended on the milling time. The Crystallinity of these samples should also be determined to decide on their suitability to be used in various industrial processes [21]. Figure 3.12 shows X-ray Diffraction (XRD).

Particle size and crystalline size are different and usually be misinterpreted. A particle may be made up of several different crystallites. Crystallite size often matches grain size, but there are exceptions. There are three method used to determine crystalline sizes which are:-

- **Scherer** method (using FWHM) gives the ratio of the root-mean-fourth-power to the root-mean-square value of the thickness

- **Stokes and Wilson** method (using integral breadth) determines the volume average of the thickness of the crystallites measured perpendicular to the reflecting plane

- **The variance methods** give the ratio of the total volume of the crystallites to the total area of their projection on a plane parallel to the reflecting planes

However, for this project, in order to determine the crystalline size, the Scherer method is calculated using MDI Jade 5.0 software. Jade is a powder diffraction analysis package. Data is collected on the diffractometer using PC-based Datascan software by MDI and processed using Jade software. The software offers a wide range of analysis functions for both qualitative and quantitative analysis of polycrystalline materials. For this project, qualitative analysis is used to determine quartz crystalline structure whereas for quantitative analysis is used to determine crystalline size.

The Scherer formula is:-

$$D = \frac{K\lambda}{\beta \cos\theta} \qquad \text{.....(Equation 3.1)}$$

Where 'λ' is wave length of X-Ray (0.15406 nm), 'β' is FWHM (full width at half maximum), 'θ' is the diffraction angle and 'D' is particle diameter size or crystalline size. The constant of proportionality, K (the Scherer constant) depends on the how the width is determined, the shape of the crystal, and the size distribution.
The most common values for K are:

- 0.94 for FWHM of spherical crystals with cubic symmetry
- 0.89 for integral breadth of spherical crystals w/ cubic symmetry
- 1, because 0.94 and 0.89 both round up to 1

(K actually varies from 0.62 to 2.08)

However, for this project the K value is taken as 0.94 since the quartz is assumed spherical crystals with cubic symmetry. The factor that effects K are:-

- how the peak width is defined
- how crystallite size is defined
- the shape of the crystal
- the size distribution

Figure 3.12: X-ray Diffraction (XRD)

3.4.4 Chemical Composition

The chemical composition of silica sand can be analysed using X-Ray Fluorescent. XRF works when a sample is illuminated by an intense X-ray beam, some of the energy is scattered, whereas some is absorbed within the sample in a manner that depends on its chemistry [19]. The intense X-ray beam is known as the incident beam [18]. X-ray fluorescence (XRF) analysis is a fast, non-destructive and environmentally friendly analysis method with very high accuracy and reproducibility [9]. Figure 3.13 shows X-Ray Fluorescent (XRF).

Figure 3.13: X-Ray Fluorescent (XRF)

3.4.5 Elemental Mapping

In addition to secondary electrons imaging, Energy Dispersive X-ray (EDX) Analysis is also useful tools widely used for chemical analysis. The intensity of backscattered electrons generated by electron bombardment can be correlated to the atomic number of the element within the sampling volume. Hence, qualitative elemental information can be revealed. The characteristic X-rays emitted from the sample serve as fingerprints and give elemental information of the samples including semi-quantitative analysis, quantitative analysis, line profiling and spatial distribution of elements. On the surface were the silicone-based water repellents were applied, thick silicone-layers were partly observed. When thick silicone-layers were visible, EDX mapping could confirm that this structure contained Si elements.

EDX was chosen for this project because it is easiest to use in conjunction with electron microscopy and it can be used to determine the composition of small particles or areas within a sample, as well as the bulk composition. Most of the other techniques do not have this ability and generally only produce information about the bulk sample, which is not useful for highly inhomogeneous samples like the sand.

The major disadvantage of EDX is that light elements (atomic number less than 11) produce very few characteristic X-rays but produce mainly Auger electrons instead. [26]. Therefore, it is difficult to obtain an accurate composition for these elements. The EDX results for element Si will be compared with results from both XRD for quartz and XRF for SiO_2. Figure 3.14 shows Energy Dispersive X-ray (EDX).

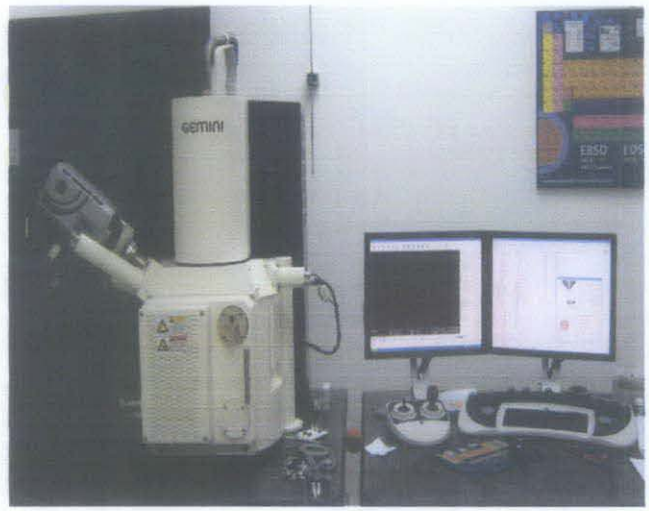


Figure 3.14: Energy Dispersive X-ray (EDX)

Table 3.4

Final Year Project 1 – Gantt Chart

No	Details/Weeks	1	2	3	4	5	6		7	8	9	10	11	12	13	14	15
1	Literature Review																
2	Methodolgy																
	2.1 Pre - Experiment																
	2.1.1 Collect silica sand																
	2.1.2 Weight 1kg silica sand																
	2.1.2 Mesh silica sand to 600um																
	2.1.3 Weight silica sand left																
	2.2 Process Preparation																
	2.2.1 Get approval to use oven																
	2.3 Technical Experiment																
	2.3.1 Prepare slurry																
	2.3.2 Grind silica sand by wet method using ball milling																
	2.3.3 Dry silica sand on oven																
	2.3.4 Mesh silica sand using Sieve Shaker																
	2.3.5 Weigh silica sand left																
3	Results / Characterizations																
	3.1 Process Preparation																
	3.1.1 Obtain form & book for XRD & FESEM																
	3.2 Characterize Results																
	3.2.1 Characterize results using XRD & FESEM																
4	Reporting																
	4.1 Prelimanary Report																
	4.2Progress Report																
	4.3 Seminar																
	4.4 Interim Report																
	4.5 Oral Presentation																

Lagend

Process

Suggested Milestone

Semester Break

Table 3.5

Final Year Project 2 – Gantt Chart

No	Details/Weeks	1	2	3	4	5	6	7		8	9	10	11	12	13	14	15
1	1.0 Literature Review	■	■	■	■	▲	■	■		■	▲	■	▲	▲	▲	▲	▲
2	2.0 Methodolgy																
	2.1 Pre - Experiment																
	2.1.1 Collect silica sand	■	▲														
	2.1.2 Weight 1kg silica sand	■	▲														
	2.1.2 Mesh silica sand to 600um	■	▲														
	2.1.3 Weight silica sand left	■	▲														
	2.2 Process Preparation		▲														
	2.2.1 Get approval to use oven		▲														
	2.3 Technical Experiment																
	2.3.1 Prepare slurry			■	▲												
	2.3.2 Grind silica sand by wet method using ball milling			■	▲												
	2.3.3 Dry silica sand on oven			■	▲												
	2.3.4 Mesh silica sand using Sieve Shaker			■	▲												
	2.3.5 Weigh silica sand left			■	▲												
3	3.0 Results / Characterizations																
	3.1 Process Preparation																
	3.1.1 Booking for Particle Analyzer, FESEM, XRD & XRF			▲													
	3.2 Characterize Results																
	3.2.1 Characterize results using Particle Analyzer, FESEM, XRD & XRF				■	■	■	▲									
4	4.0 Report & Presentation																
	4.1 Progress Report						■	■		■	▲						
	4.2 Pre-EDX									■	■	■	▲				
	4.3 Draft Report											■	■	▲			
	4.4 Dissertation (Soft Bound)												■	■	▲		
	4.5 Technical Paper												■	■	▲		
	4.6 Oral Presentation													■	■	▲	
	4.7 Dissertation (Hard Bound)														■	■	▲

Lagend

■ Process

▲ Suggested Milestone

Semester Break

CHAPTER 4
RESULTS & DISCUSSION

4.1 Sample Observation

Figure 4.1 below shows silica sand fine particles obtained after 6 hours of grinding.

Figure 4.1: Silica sand fine particles

4.2 Mesh Size using Sieve Analysis

The Table 4.1 shows that the quantity of silica sand used at 1st stage is 100 g. For simplicity, the quantity of silica sand is presumed to be 100 g for subsequent stages even though the actual quantity has slightly reduced since some of them used as sample for characterization using advanced equipment. Furthermore, some of silica sand which was larger size from the pan size was wasted during the sieve analysis. However, all these wasted sand is assumed neglected.

Using sieve analysis, it shows that the size of silica sand particles were reduced sequentially (<425 μm, 212 μm, 150 μm and 63 μm and fine particles). By wet method approaches, silica sand particles was ground down until getting size less than 63μm in 4 hours. The silica sand particles continued to be ground about two hours in order to obtain very fine particle of silica sand.

The lowest pan size of sieve shaker is 63 μm. Thus, one of the limitations of using sieve analysis is due to its limited meshing size. By four hours of grinding duration, the silica sand particles sizes are already reduced to less than 63 μm. Thus, it is not practical anymore to use sieve shaker after size less than 63 μm. Figure 4.1 below illustrates the meshing sizes of sieve shaker versus grinding duration.

Table 4.1: Sieve Analysis of Wet Method Mechanism

Stage	**Parameters**					**Sieve Analysis on Particles Size (μm)**
	Silica Sand Quantity (g)	Grinding Speed (rpm)	Grinding Duration (hour)	Bead Quantity (pcs)	Water Quantity (ml)	
1st	100	90	1	320	60	<425
2nd	100	90	1	320	60	<212
3rd	100	90	1	320	60	<150
4th	100	90	1	320	60	<63
5th	100	90	2	320	60	<63

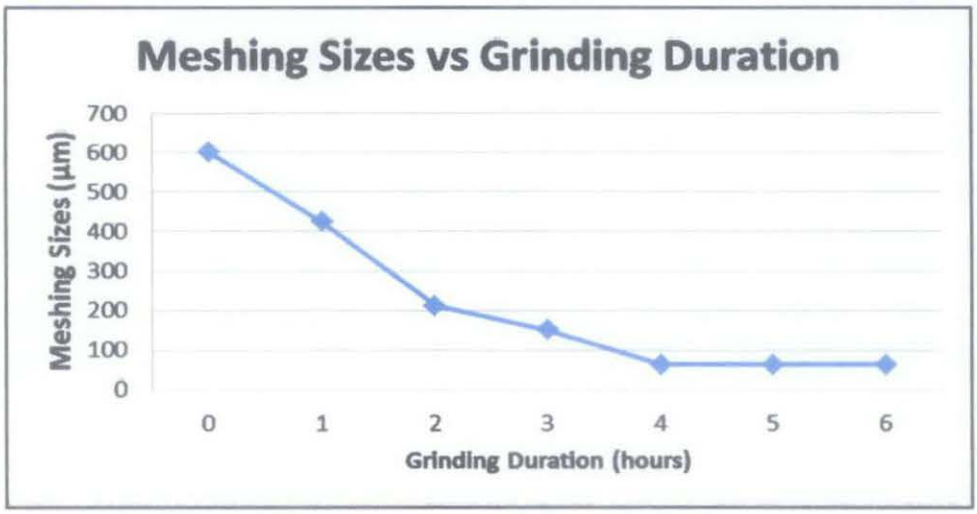


Figure 4.2: Graph of meshing size versus grinding duration

4.3 Particle Size Distribution

The particle size distribution of silica sand was recorded using Mastersizer 2000 particle size analyzer. The particle size distribution of silica sand is shown on Figure 4.3. The graph shows the volume percent as a function of particle size. The green line shows the initial size distribution of silica sand particles at 0 hours of grinding. Meanwhile, the red colour shows the particle size distribution of silica sand at 6 hours of grinding.

It could be seen that the distribution for both graphs were monomodal and bimodal respectively, which allowed the use of volume moment diameter to characterize the results and consequently to define the fineness criteria based on volume moment diameter. It indicates from the graph that the monomodal of particle size distribution become bimodal as grinding time is increased and the peak at lower particle size emerged when the analysis were carried out on volume basis.

There were four sub-populations noticed from the graph. For 0 hour of grinding, particle size distribution is centered at about 300 μm while for lower particles size, centered between 5 μm and 100 μm. However, after 6 hours of grinding, the particle size distribution centered at about 5 μm while lower particles size centered between 0.5 μm and 1 μm. Thus, in can be concluded that the size distribution of silica sand particles decreased as the grinding time is increased.

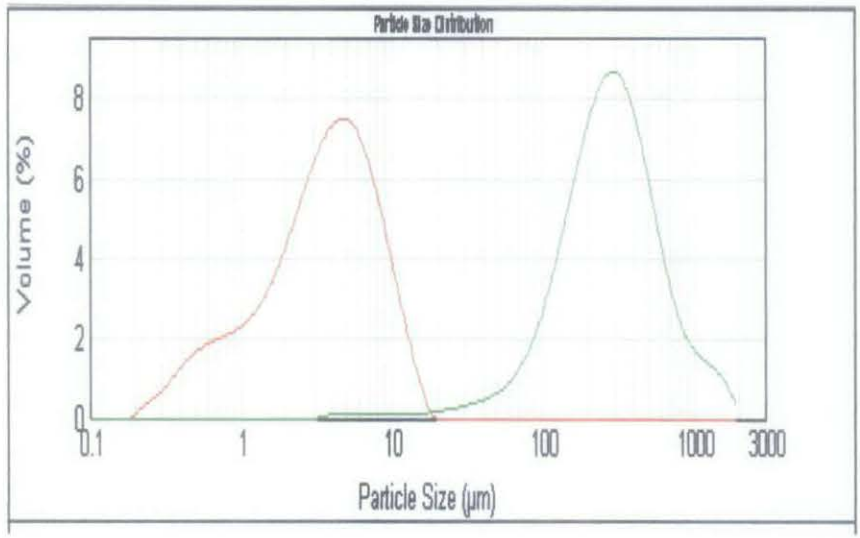


Figure 4.3: Particle size distribution of silica sand

4.4 Morphology Evolution

In the beginning, Scanning Electron Microscope (SEM) characterization was done to the silica sand particles by using 1KX of magnification level. However, it is difficult to see the exact shape of the particles at higher magnifications as the image is blurred. Basically, SEM analysis can go even greater of its magnification level than 1KX, but it takes longer time to render and load the images. Therefore, Field Emission Scanning Electron Microscope (FESEM) characterization was carried out at 20KX.

Morphological evolution of silica sand powders during grinding at every hour was investigated both for SEM and FESEM at different resolutions. Figure 4.4 shows SEM micrograph of silica sand at different grinding durations. As can be seen, the silica sand particles at 0 hour of grinding are too close thus difficult to determine the exact shape of the particles since agglomeration is too high. As the grinding hour increased to 2 hours, 4 hours and 6 hours the particles shape is much easier to be determined but the images are still blurred.

To see the silica sand particles image at smaller field of view, higher magnification of image is being used. The higher the magnification, the better the resolution is thus the more crisp the image is. Field Emission Scanning Electron Microscope (FESEM) is used for higher magnification with magnification of 20KX. As can be seen from Figure 4.5, the image shows different micrograph of silica sand particles at 20KX with different grinding durations.

At 0 hour of grinding as shown from Figure 4.5 (a), the image shows semi-globular, flaky and fine shape (irregular shape) of silica sand particles. The particles are partially agglomerated, but generally have fine edges. As the grinding duration is increased to 2 hours as shown from Figure 4.5 (b), most of the silica sand particles have flaky and fines shape with smaller in size. It should be mentioned that the local temperature caused by ball collision could be much higher than the silica sand temperature. At this temperature, the silica sand chains have enough mobility. Therefore, at this stage, the silica sand particles got flattened and deagglomerated.

Continuous grinding to 4 hours caused silica sand particle to be away from each other as shown from Figure 4.5 (c). This is due to repetitive impacts of grinding media with silica sand particles and severe drying of silica sand in oven with smaller contents of silica sand but at same temperature. Less agglomeration can be seen.

By further grinding up to 6 hours as shown from Figure 4.5 (d), the size of silica sand particles is reduced and this silica sand particles show that they are more away towards each other. Agglomeration of silica sand particles still exists but with less agglomeration. The small white points being observed could be related to silica sand particles agglomerations.

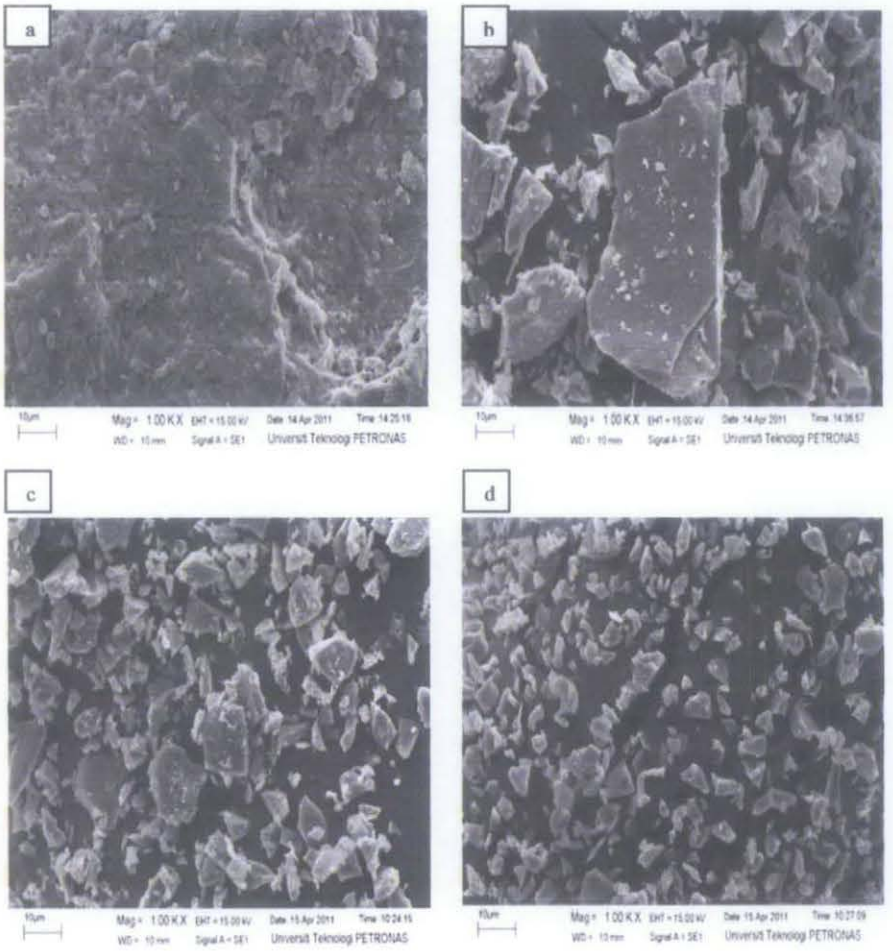


Figure 4.4: SEM micrographs of silica sand at 1KX with different grinding durations (a) 0 hour, (b) 2 hours, (c) 4 hours and (d) 6 hours

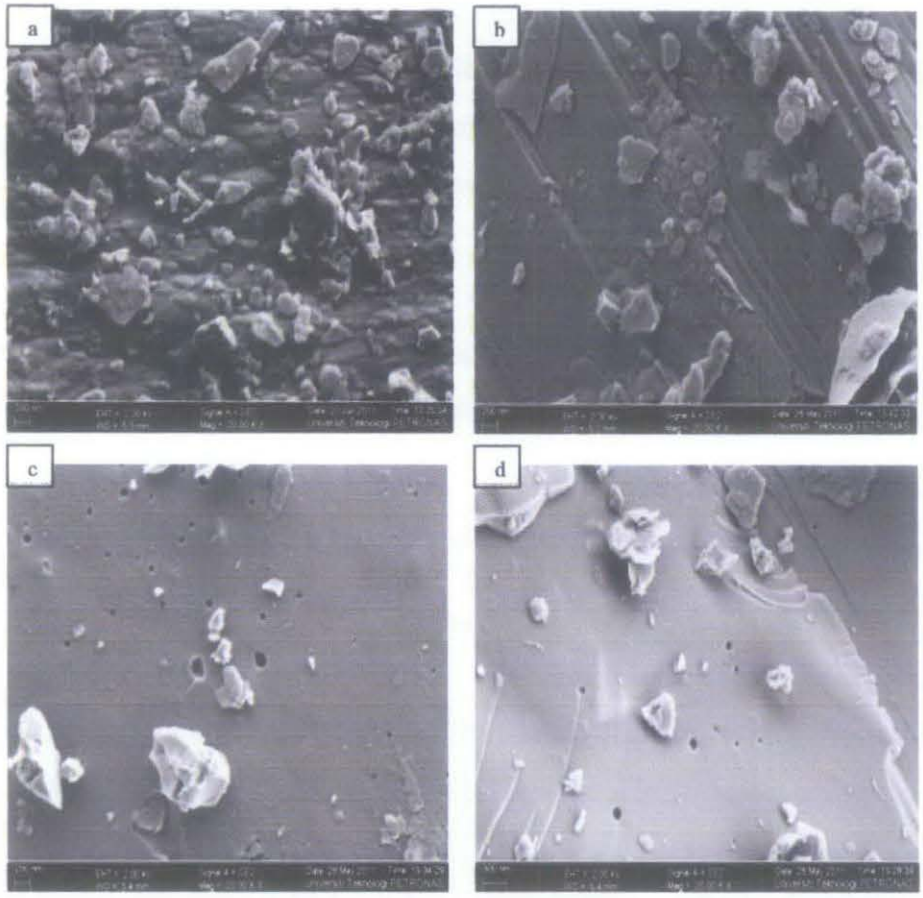


Figure 4.5: FESEM micrographs of silica sand at 20KX with different grinding durations (a) 0 hour, (b) 2 hours, (c) 4 hours and (d) 6 hours

4.5 Crystalline Structure and Size

The corresponding diffractograms to the lattice planes at different sizes are indicated. The lattice plane [110] indicates the strongest line. Full Width at Half Maximum (FWHM), the width of the diffraction peak, in radians, at a height half-way between background and the peak maximum. The initial sharp peaks of quartz were broadened due to the refinement of the crystalline size and the generation of strain with increasing grinding duration. As the intensity of the quartz peaks decreases with increasing of grinding duration, the positions of these peaks were unchanged, indicating the possible dissolution of quartz. It also said that even after 6 hours of grinding, the characteristic X-ray diffraction peaks of quartz were still observable.

It was concluded that the crystalline structure could be partly retained even after quite long grinding duration. Thus, the XRD result shows that as grinding duration is increasing, there is no transformation of crystalline structure of quartz to amorphous. The XRD pattern at different grinding duration is shown in the Figure 4.6

From this study, considering the peak at degrees, average crystalline size has been estimated by using Scherer formula obtained from MDI Jade software. At highest peak plane, the crystalline size is determined using Scherer Formula below:-

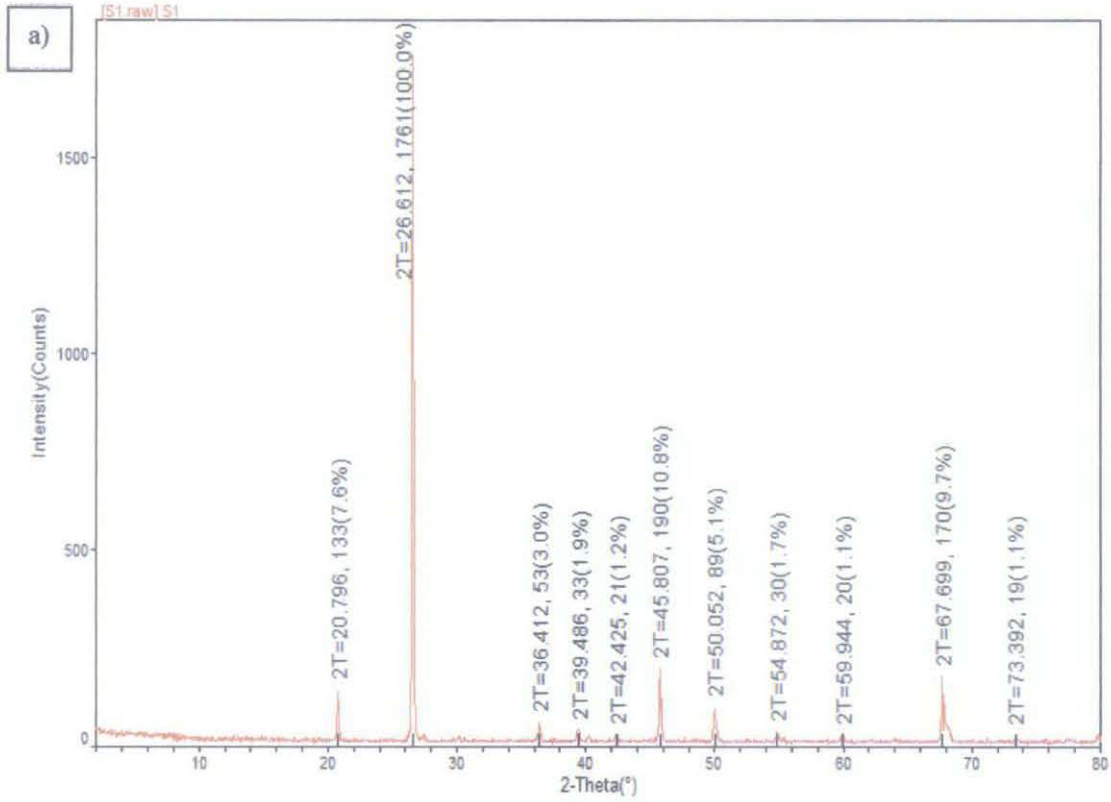
$$D = \frac{K\lambda}{\beta \cos\theta}$$

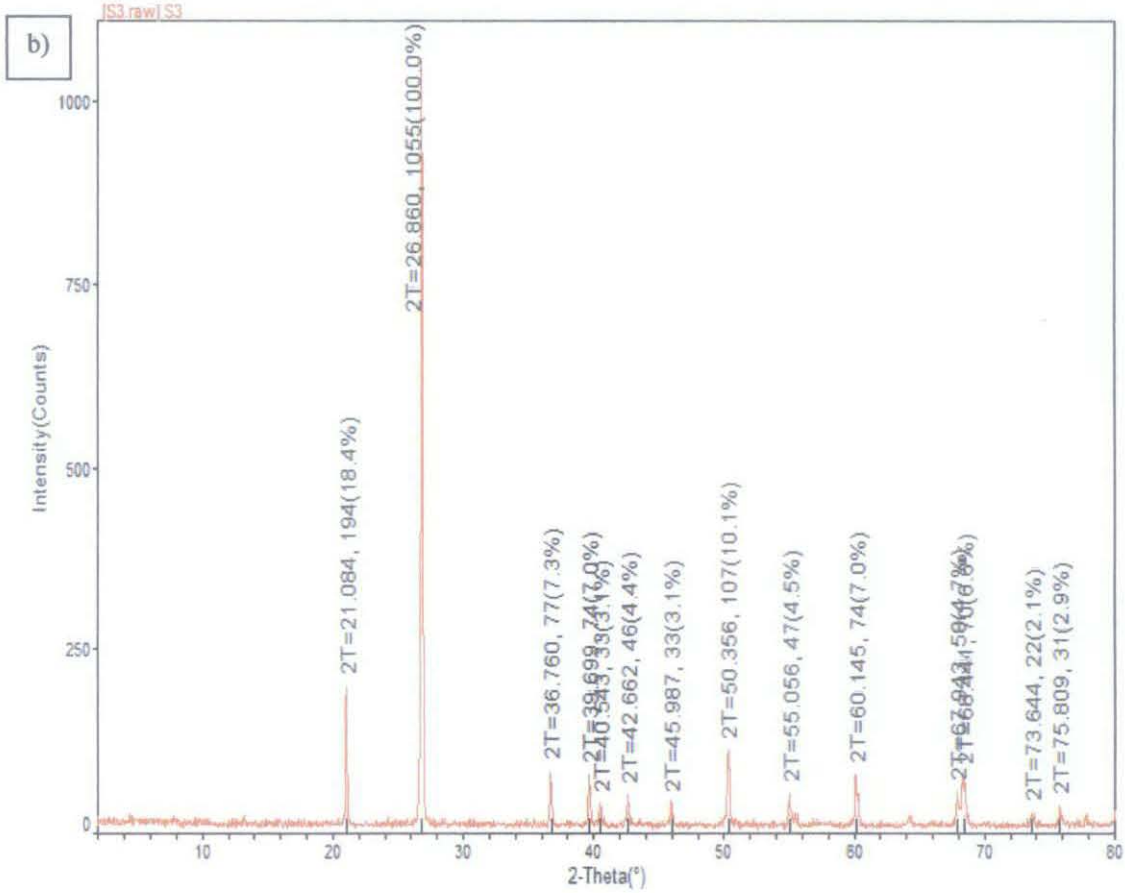
.....(Equation 4.1)

Where 'λ' is wave length of X-Ray (0.15406 nm), 'β' is FWHM (full width at half maximum), 'θ' is the diffraction angle and 'D' is particle diameter size or crystalline size. K is Scherer constant by taking K=0.94. Table 4.2 shows that at highest peak plane of Miller Indices [1 1 0], as the grinding duration is increased, the crystalline size of quartz is decreased with decreasing of quartz intensity. Figure 4.7 shows the quartz intensity decreases as grinding duration increases. Meanwhile, Figure 4.8 shows the crystalline size of quartz decreases as grinding duration increases. Figure 4.9 illustrates the combination of XRD results at different grinding durations

Table 4.2: XRD results for quartz at different grinding duration

Grinding Duration (hours)	Highest peak plane for Miller Indices [hkl]	Diffraction angle, 2θ	Intensity (Counts)	Crystalline size, D (nm)
0	[110]	26.612	1761	624
2	[110]	26.860	1055	425
4	[110]	26.798	1020	400
6	[110]	26.893	684	347


a)
[S1.raw] S1
Intensity(Counts)
2-Theta(°)
2T=20.796, 133(7.6%)
2T=26.612, 1761(100.0%)
2T=36.412, 53(3.0%)
2T=39.486, 33(1.9%)
2T=42.425, 21(1.2%)
2T=45.807, 190(10.8%)
2T=50.052, 89(5.1%)
2T=54.872, 30(1.7%)
2T=59.944, 20(1.1%)
2T=67.699, 170(9.7%)
2T=73.392, 19(1.1%)


b)
[S3.raw] S3
Intensity(Counts)
2-Theta(°)
2T=21.084, 194(18.4%)
2T=26.860, 1055(100.0%)
2T=36.760, 77(7.3%)
2T=42.662, 46(4.4%)
2T=45.987, 33(3.1%)
2T=50.356, 107(10.1%)
2T=55.056, 47(4.5%)
2T=60.145, 74(7.0%)
2T=73.644, 22(2.1%)
2T=75.809, 31(2.9%)

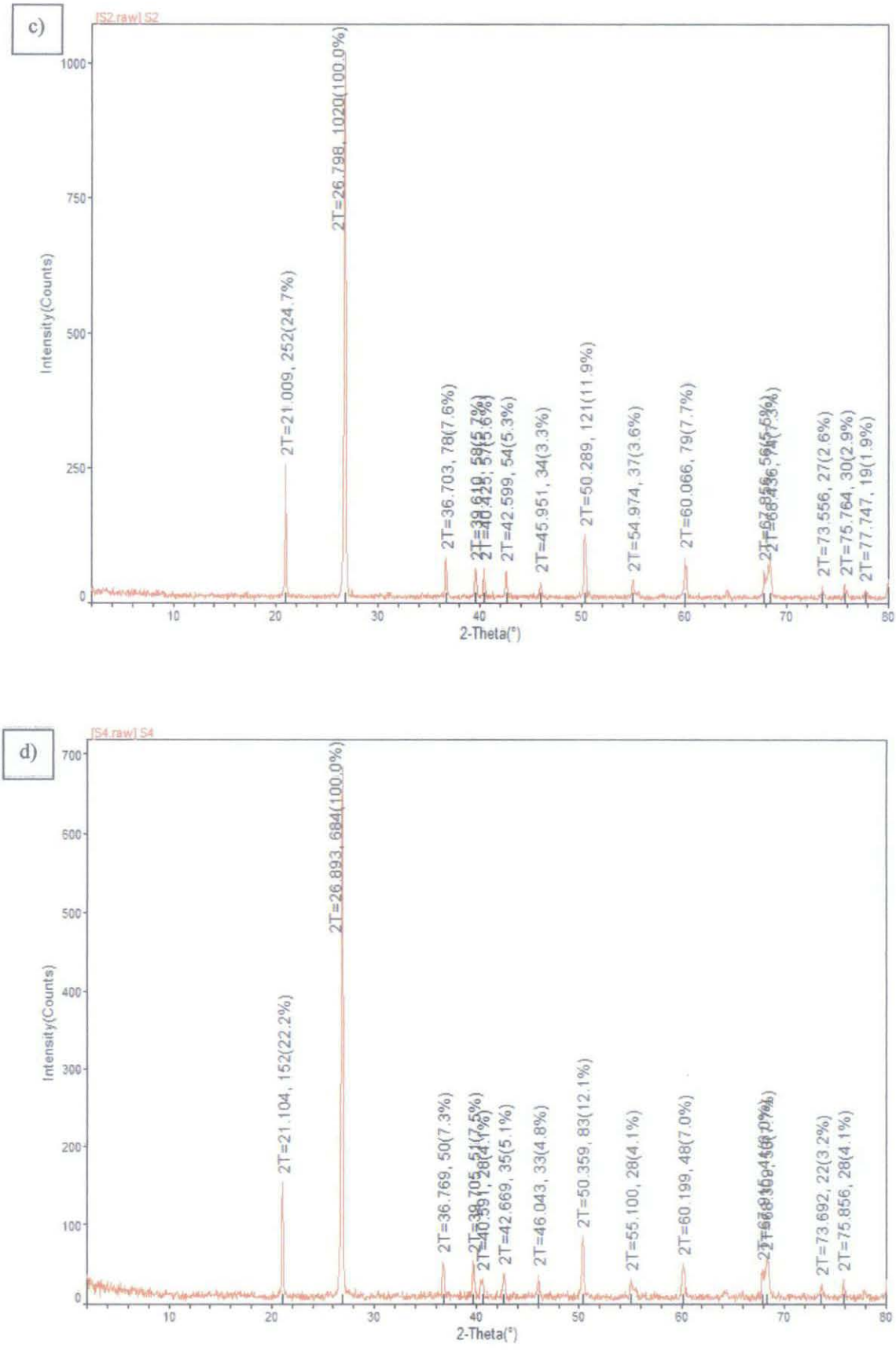


Figure 4.6: XRD results at different grinding duration a) 0 hour b) 2 hours c) 4 hours and d) 6 hours

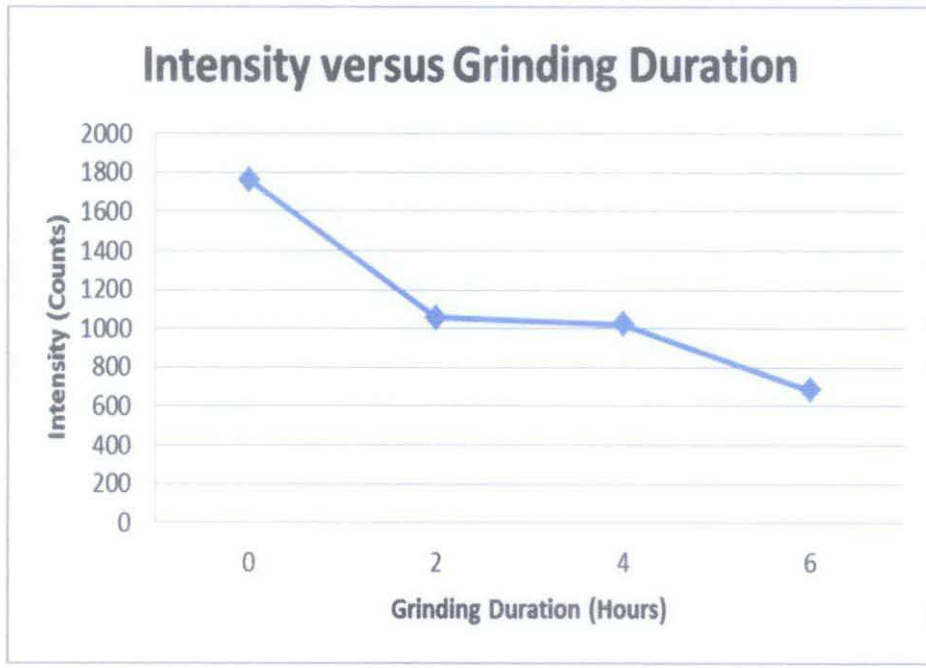


Figure 4.7: Intensity versus grindning duration

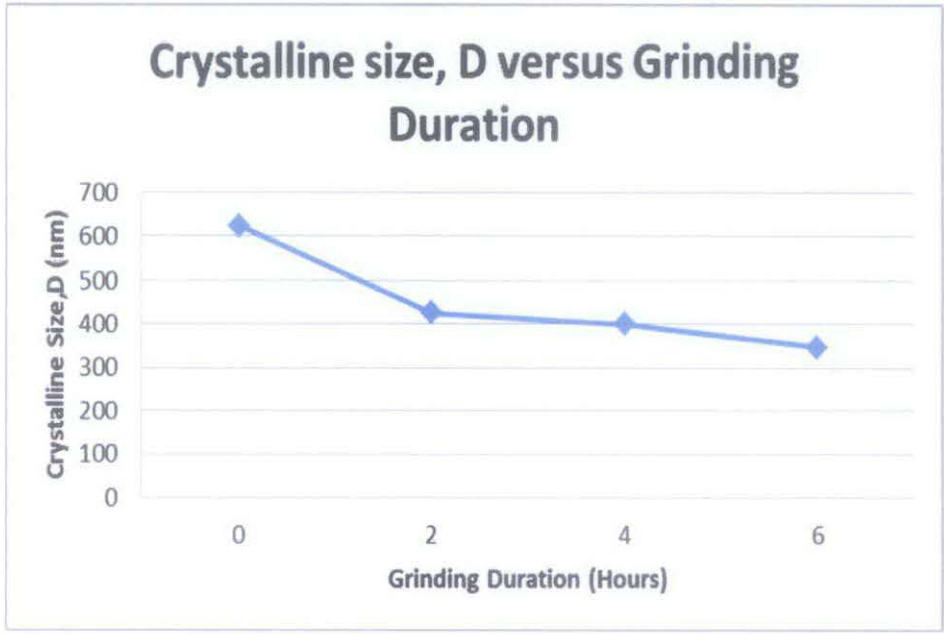


Figure 4.8: Crystalline size, D versus grinding duration

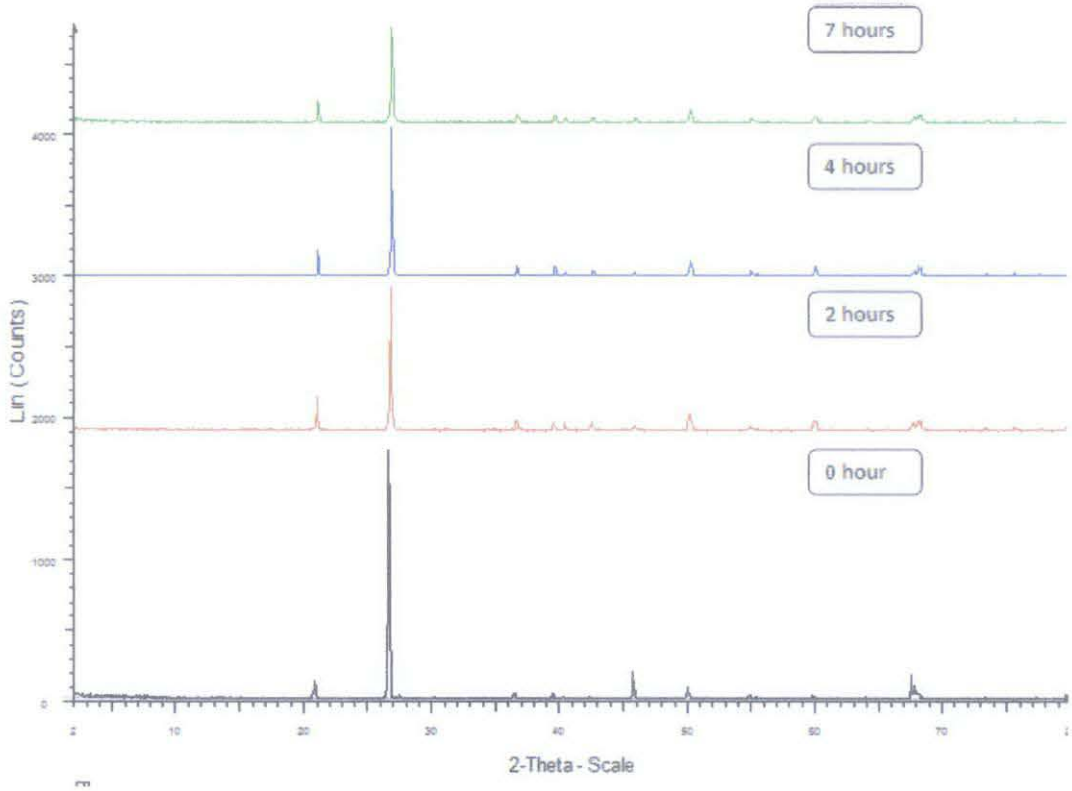


Figure 4.9: Combination of XRD results at different grinding durations

4.6 Chemical composition

X-Ray Fluorescence (XRF) is done to determine the chemical composition of silica sand. Samples were undergoing the X-Ray Fluorescence (XRF) analysis as to reveal the composition of oxides of silica sand at different grinding duration.

The data given in Table 4.4 shows that both silicon dioxide and alumina are present in major quantities while other compositions are present in trace amount. Other compositions are impurities and assumed negligible. Based on the XRF analysis, Tronoh raw silica sand has the chemical composition of SiO_2 of 89.9%. The composition of SiO_2 tends to be increased as the grinding hours increased (Figure 4.10). After 6 hours of grinding, the composition of SiO_2 is 96.5%. The result shows high content of silica and proved by I. Saaid, D. Kahmat & S. Muhammad [25].

Meanwhile, Figure 4.10 also shows decreasing amount of Al_2O_3, from 9.9% to 3.2% as grinding duration increased. It shows that composition of Al_2O_3 is dissolved as grinding duration increased. Table 4.3 illustrates the relationship between the grinding duration and silica concentration at different hours of grinding (0 hour, 2 hours, 4 hours, and 6 hours).

Table 4.3: Chemical Composition of silica sand

Grinding Duration (Hours) / Oxides (%)	**Quantity of SiO_2 (%)**	**Quantity of Al_2O_3 (%)**
0	89.90	9.900
2	95.00	4.470
4	96.50	3.200
6	96.50	3.200

Notes: Other chemical composition are impurities and assumed negligible

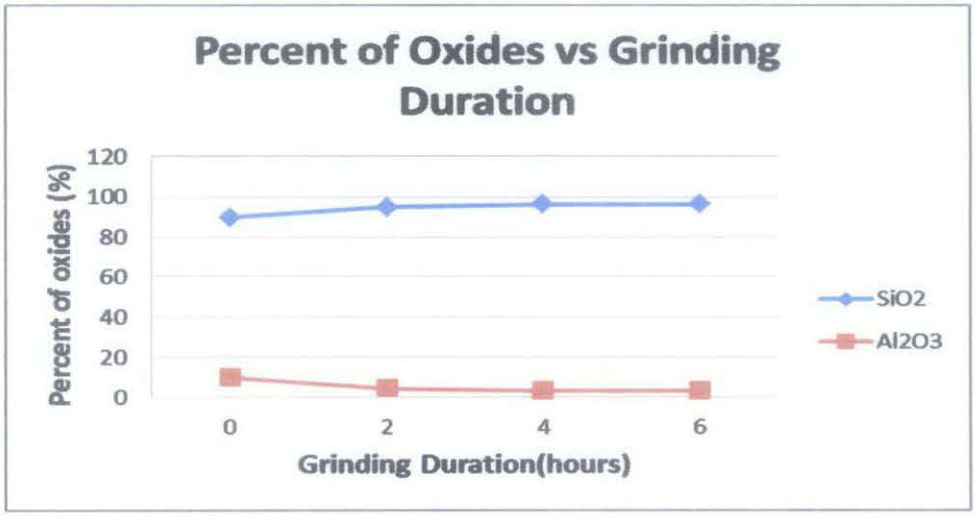


Figure 4.10: Percent of oxides versus grinding duration for both SiO_2 and Al_2O_3

4.7 Elemental Mapping

Figure 4.12 shows the result from mapping using Energy Dispersive X-ray (EDX) at different grinding duration. From Table 4.4, as the grinding duration is increased, the element of Si for both weights (%) and atomic (%) are decreased. The signals for Si element correlated with the location of quartz crystals from XRD result. Figure 4.11 illustrate the element of Si weight (%) and atomic (%) vs grinding duration. This result supports the XRF result where show that as grinding duration is increased, the composition of SiO_2 is increased.

Table 4.4: EDX mapping at different grinding duration

Grinding Duration (Hours)	Si		Al		O	
	Weight %	Atomic %	Weight %	Atomic %	Weight %	Atomic %
0	32.92	21.42	2.62	1.78	55.33	63.19
2	30.88	19.30	-	-	55.81	61.24
4	27.03	15.70	-	-	43.65	44.50
6	26.29	15.49	-	-	49.56	51.25

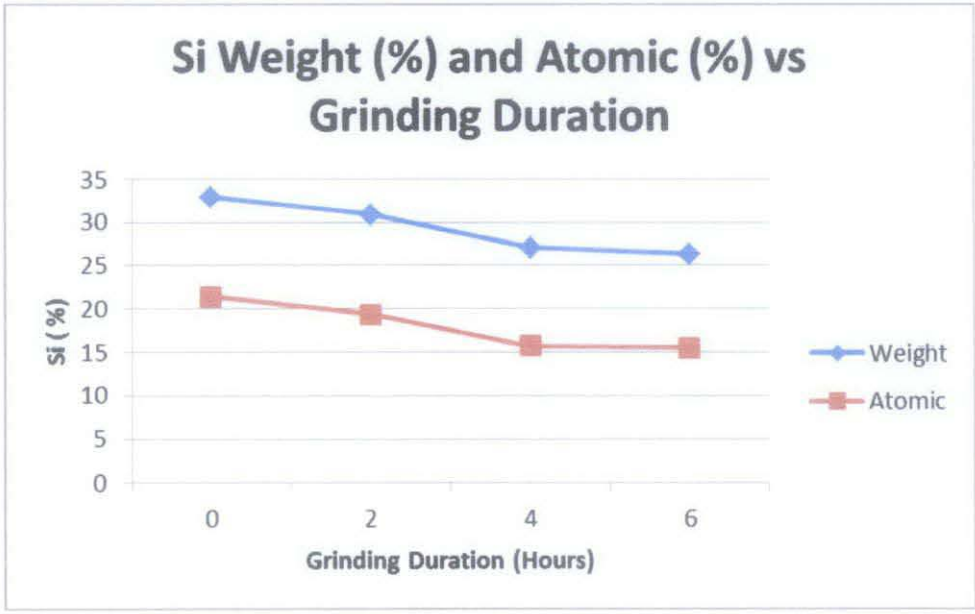


Figure 4.11: Si weight (%) and atomic (%) vs grinding duration

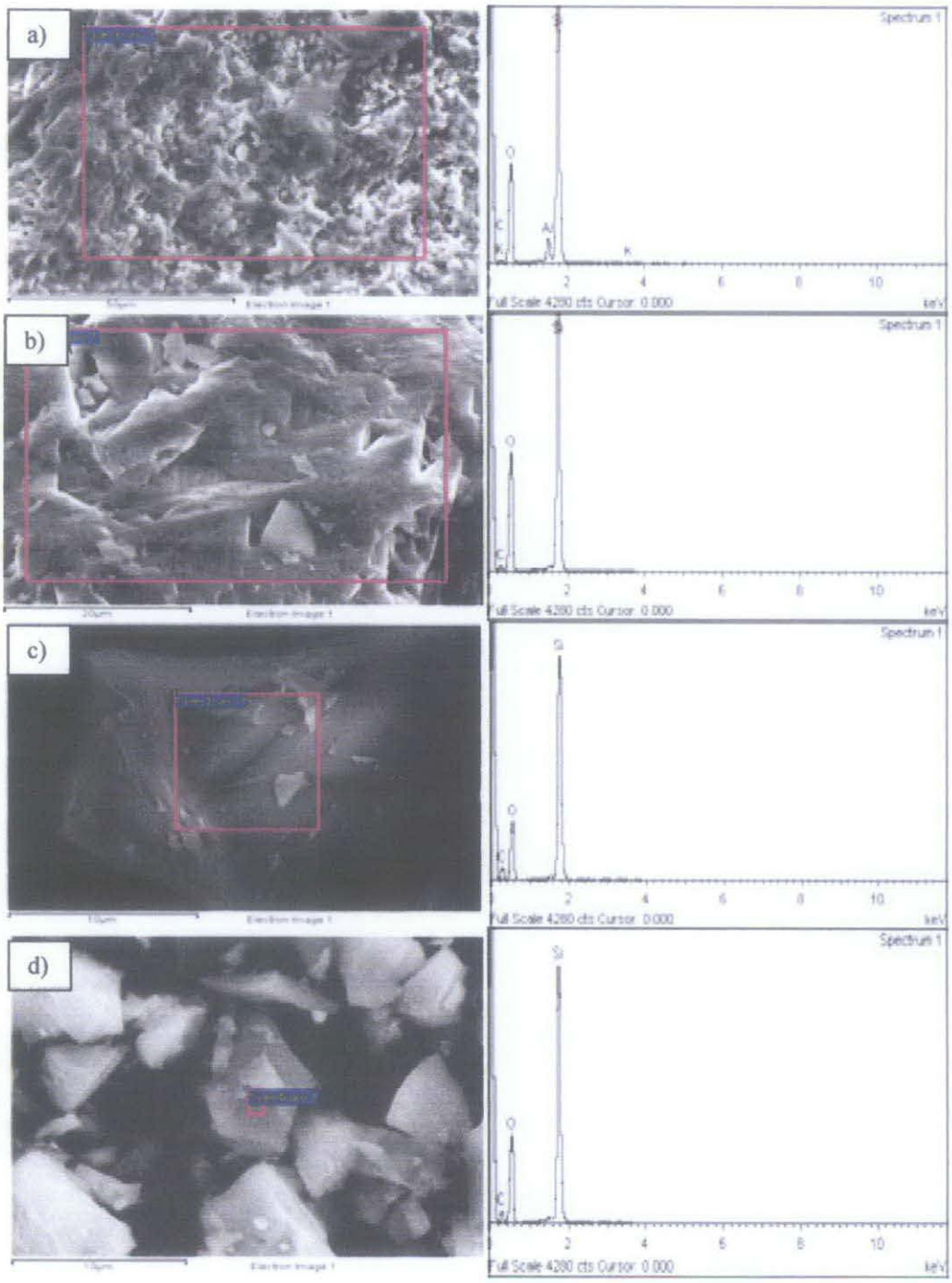


Figure 4.12: EDX mapping of silica sand at different grinding duration a) 0 hours b) 2 hours c) 4 hours and d) 6 hours

CHAPTER 5
CONCLUSION AND RECOMMENDATION

5. 1 Conclusion

After doing all the research and experiment regarding this project, ball mills approach has proved its capabilities to produce silica sand fine particles by using wet milling method. Based from the results gathered in Chapter 4, US STONEWARE 764AVM Ball Mill machine is capable to produce silica sand fine particles with less than 63 μm after six hours of grinding time. Using sieve analysis, the size of silica sand particles can be reduced to size <63 μm after 4 hours of grinding. The particle size distribution of silica sand can be determined by using Mastersizer 2000 particle size analyzer. It shows that as the grinding duration is increased, the smaller size distribution of silica sand particles can be obtained. The morphology evolution of silica sand particles can be determined by using both Scanning Electron Microscope (SEM) and Field Emission Scanning Electron Microscope (FESEM). As grinding duration increased, SEM shows blurred image while FESEM shows reduction of silica sand particles size and the changes in morphology of silica sand from irregular shapes particles to particles with flaky and irregular shape which are more away towards each other. The crystalline structure of silica sand can be determined by using X-Ray Diffraction (XRD). It shows that the structure of silica sand remains crystalline as grinding duration is increased. Meanwhile, at highest peak plane of Miller Indices [1 1 0], as the grinding duration is increased, the crystalline size of quartz is decreasing with decreasing of quartz intensity. From X-Ray Fluorescence (XRF) result, it shows that as grinding duration is increased, the concentration of SiO_2 increased. The result is supported from Energy Dispersive X-ray (EDX) result where shows decreasing element of Si as grinding duration is increased.

5.2 Recommendation

5.2.1 Grinding media

The rate of grinding depends on several factors. One of the factors is to use grinding media. In this case, grinding media that being used is zirconium ceramic beads. In order to increase the rate of grinding, the grinding media should consists of material with as high density as possible [20]. The size of grinding media also is an important consideration. Small grinding media are generally better that large ones [20]. It is said that as the ball diameter is decreased, finer particles may be expected to be obtained [21].

Moreover, the particle size distribution width was lowered by using smaller grinding balls in wet condition and larger grinding balls in dry condition [22]. Assuming that the rate of grinding depends on the number of contact points between the balls and the powder and that the number of contact points in turn depends on the surface area of the ball. Then, the rate of grinding will increase inversely as the radius of the balls. However, the balls cannot be too small because they must impart sufficient mechanical energy to the particles to cause fracture.

5.2.2 Use of steel balls

The disadvantage of ball milling is that wear of the grinding can be fairly high. For advanced ceramics, the presence of impurities in the powder is serious problem. The best solution is instead of using zirconium ceramic beads as the mill ball, we can use steel balls [20]. By using steel balls, the ball can be easily removed from the powder, thus avoid any contaminations. It must be noted that the use of porcelain balls or low purity Al_2O_3 balls must be avoided because these ball can wear easily and thus introduce a fair amount of SiO_2 into the powder.

5.2.3 Particle size

The rate of grinding also depends on the particle size. The rate decreases with decreasing particle size and as the particle become fairly fine, it becomes more and more difficult to achieve size reduction. Thus, to solve this solution, a practical grinding limit is done.

First, the increased tendency for the particles to agglomerate with decreasing particle size means that a physical equilibrium is set up between the agglomeration and comminution processes.

Second, the probability for the occurrence of a comminution event decreases with decreasing particle size.

Third, the probability of a flaw with a given sizes existing in the particle decreases with decreasing particle size. The limiting particle size may be made smaller by wet milling as opposed to dry milling, the use of a dispersing agent during wet milling (e.g. Flotigam P, an organic dispersing agent), and by performing the milling in stages [20].

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