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Research article

Particle Morphology and Optical Property of Silicate Powder Prepared from Waste Borosilicate Glass by High Energy Ball Milling Process

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Abstract

The aim of this work was focused on the size reduction of waste glass that was obtained from broken beaker glass in the laboratory. The particle size reduction of glass cullet was performed by a high energy ball milling process. Firstly, the obtained beaker glass was smashed into fine particles before the milling process. The milling parameters studied, which had potential effects on the efficiency of the milling process, were rotation speed, milling time and ball sizes. The particle size and morphology of before/after milled silicate powder were evaluated by a particle analyzer and a field emission scanning electron microscope (FE-SEM). It was shown that the particle size of the milled powder had decreased after milling. However, agglomeration occurred after the use of high rotation speeds and longer milling times. Furthermore, functional groups of the milled samples were analyzed by a Fourier transform-infrared (FT-IR) spectrometer. The FTIR spectra of samples of each milled condition displayede similar patterns. Meanwhile, an optical property, the scattering effect of milled samples of different particle sizes, was identified by a luminance meter. The preliminary results suggested that the minimum particle size of the milled glass was reached at 1.33 μm with milling time of 30 min at 500 rpm. Therefore, an increase of light scattering property occurred, and this correspondented to the smaller particle size of milled silicate powder.

Keywords

milling process;
operation time;
rotation speed;
size reduction;
wasted glass

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1. Introduction

Waste glass materials destined for recycling processes typically include three types of glass waste: perfect, unbreakable or damaged glass. However, glass materials that are in a broken form are not suitable for recycling and are usually disposed of as waste. This is considered to be a crucial environmental problem in Thailand. In laboratory, a beaker is a clear glass vessel with a cylindrical shape and flat bottom, and a beak or "spout". Beakers are some of the most important pieces of equipment used in research. They are used to heat and mix chemical solutions, and chemical reactions are often performed in them. Beakers range in volume from 50 to 5,000 ml, and are usually made from borosilicate 3.3. Beakers display a range of special features including inertness and possess high chemical resistance and minimal thermal expansion. As a result of their resistance to thermal shock, they can be used at high temperature [1]. When a beaker is damaged or broken, it must be disposed of as useless waste. Then, waste beakers are of worthy of study for their potential as scattering materials because of their prominent physical properties and high transparency. Waste beaker glass was the focus of this work because its main component is silicon dioxide (SiO_2) or silica. The silica is mixed with other components to form borosilicate glass. One of the main unique optical and mechanical properties of silica powder is its good light penetration in the visible spectral region due to its highly transparent material [2]. Hence, the reduction of silica powder into nano-scale is of interest for light scattering applications. When silica particles are filled into light-guide plates, a difference of refractive index between silica NPs and the light-guide plate matrix occurs. The way light penetrates to the light-guide plate and hits the silica particles inside relates to the increase of light scattering inside the plate [3]. Therefore, an improvement of light intensity can be transmitted in front of the plate by the influence of scattering material. Suthabanditpong *et al*. [4] reported the improvement of diffuse transmittance of a light diffuser film by the presence of hollow silica nanoparticles. An increment of the silica particles also leads to the formation of aggregated particles with a larger size and homogeneous dispersion with the appearance of the opacity, high transmittance that are close to that of the cleaned glass. The fabrication of light diffusing films with high transmittance and haze simultaneously by mimicking the compound of polymethyl methacrylate (PMMA)/SiO_2 composite microspheres was reported by Guo *et al*. [5]. More uniform and downy LED downlight was observed with the benefit of light diffusing coating film of strawberry-like PMMA microspheres and colloidal SiO_2 nanoparticles on the PET substrate. Meanwhile, small spherical particles of the composite microspheres of polycarbonate (PC)/poly (styrene-co-acrylonitrile) (SAN)-SiO_2 as light scattering materials could produce multiple scattering effect, reducing the direct transmittance and increasing the scattering transmittance relating to greatly improve the light-scattering performance in the device. This advantage of silica particles was proposed by Ding *et al*. [6]. In this work, we focused on size reduction form beaker glass cullet as the product of silicate (SiO_4) particles. Therefore, it was a challenge to produce silicate particles sized in the micro or nanoscale range and compare their optical properties with silica particles.

Mechanical milling process is one of possible process for size reduction in nanomaterials. Dry and wet milling processes are usually the main methods used to synthesize nanomaterials from a top-down approach. Wet media milling based water or fluid phase produces physically stable nanometer-sized particles of poorly water soluble compounds. Wet milling is preferable to dry milling as it requires comparatively less energy for grinding operation [7]. The recognized draw-back of the wet media milling process is a shearing of media against the active compound and the milling chamber relating to particle size reduction [8]. Meanwhile, the selection of a suitable surfactant of milling media and nanoparticles during the wet milling operation is essential if agglomeration due to cold welding is to be avoided. For dry milling process, a high grade product at the nanometer scale was obtained using a planetary ball mill at various milling times. The synthesis of silica nanoparticles with an average particle size of 80 nm using a planetary ball mill

for 30 h was observed by Wahyudi *et al*. [9]. Therefore, a planetary ball mill became of interest in our work as we investigated size reduction from waste glass cullet. A planetary ball mill is a type of high energy milling machine that can produce ultrafine particles in the nanometer range. A planetary ball mill machine is normally composed of 2 or 4 vessels on a revolving disk that rotates around a central axis, while the vessels are simultaneously rotating around their own axes. Due to this mechanism, the large impact energies that originate inside the vessels produce an effective milling by the rotation speed of both of vessels and disk in counteractive directions. The materials under mechanical process are mashed due to the impact and frictional forces caused by collisions. Thus, materials loaded into the vessels are quickly and effectively comminuted by the impact, friction and shear forces resulting from ball-to wall and ball-to-ball collisions [10]. In the ball milling process, several different variables influence the success of each individual milling process. The crucial parameters in the milling process are focused on rotation speed, milling time, weight ratio of powder to ball, and ball size diameter. These parameters affect the efficiency of the making of the fine powder product [11].

In this work, fine powder from waste beaker cullet was prepared by a high energy ball milling process. The milling parameters, different rotation speed, milling time and diameter of ball, were the main focus of study. Morphology, chemical bonding and particle size analysis were investigated to find out the influence of these parameters. In addition, the light scattering property of milled samples with different particle sizes was studied to identify the optimized particle size obtained from waste glass culler by the mechanical milling process.

2. Materials and Methods

Broken beaker glass that was to be broken down into fine powder via the high energy ball milling process was obtained from our laboratory. Firstly, the beaker glass was smashed with a hammer to make coarse glass cullets. Then, the glass cullets and zirconia balls were loaded into zirconia containers at ratio 1:10 by weight. The tilted planetary ball mill machine was operated by Planet M2-3F. The rotation speed was varied at 300, 400, 500 and 600 rpm for 30 min, while the milling time was operated at 0, 30, 60 and 90 min at rotation speed 500 rpm. Furthermore, the diameters of balls studied in the milling process were 5 mm, 10 mm and mixed 5 and 10 mm, operating at 500 rpm for 30 min. Subsequently, the silicate powder after the milling process was investigated by various techniques. Physical morphology of before/after-milled samples was monitored using a field emission scanning electron microscope (FE-SEM; SU8030, Hitachi). The chemical functional groups of all samples were analyzed by a Fourier Transform-Infrared spectrometer (FT-IR; Spectrum Two, PerkinElmer Scientific). The average particle size of milled powder was evaluated using a particle analyzer (Delsa Nano C, Beckman Coulter). Furthermore, the light scattering properties of milled samples was detected by a lux meter (Light meter, DIGICON LX-71). The samples were prepared by mixing 0.25g of milled powder (at different milling speed) with PVA solution (3.5%v/v) in a handmade acrylic transparent container with volume 250 ml. Meanwhile, a transparent solution of each mixture was evaluated in transmission mode in a double beam UV-Vis spectrometer (T92+, PG Instruments).

3. Results and Discussion

3.1 Particle size analysis of silicate powder before/after milling process

Figure 1 shows the average particle size of milled silicate powders at different rotation speeds, milling times and ball diameters in the range of 1.3-2.0 μm compared with coarse powder. Based on the effect of milling speed, the average particle size of milled samples drastically decreased with increasing milling speed, as shown in Figure 1(a). The optimized rotation speed for silicate powder in the high energy milling process was found to be 500 rpm, which produced the smallest average particle size of 1.33 μm. The size reduction of glass cullet occurred due to the effect of the strong impacts between balls and material at high rotation speed in the milling process. Moreover, fracture and collision forces between balls and material were enhanced by high milling speed related to the increase of particle size. For low rotation speed, the impact force of ball movement in the system was not enough to hit on the material structure, resulting in larger particle size from the deteriorated milling efficiency. Meanwhile, agglomeration of particles occurred at rotation speeds over 500 rpm owing to the accumulation of high thermal energy in the milled container. The second milling parameter is the effect of milling time as shown in Figure 1(b). An effective milling process at milling speed 500 rpm occurred at milling time of 30 min, which caused size reduction of silicate powder to the smallest particle size. For prolonged milling time, the average particle size tended towards high aggregation due to the enhancement of fused particles surrounded by other particles. This was an increase of particle size [12]. In addition, ball diameter was another crucial parameter in the milling process due to the impact on material as depicted in Figure 1(c). The smallest particles were significantly obtained at ball diameter of 5 mm. Meanwhile, an increase of particle size occurred when using ball diameter at 10 mm. In this case, high impact was promoted by the operation with ball diameter of 10 mm, however, some voids between the balls played a key role in the milling process because the material in the voids could not be smashed by other balls, effectively retarding milled efficiency [13]. In case of mixing balls of diameters 5 mm and 10 mm, the milled glass particles were the same size as those of the sample milled with only balls of diameter 5 mm. This condition was chosen to solve the problem of the void between the 10 mm diameter balls; however, the void cavities should be less than the diameter of 5 mm in order to avoid low milling efficiency due to the voids between 10 mm balls. Therefore, the smallest particle size of silicate powder in this work was clearly produced at rotation speed 500 rpm with milling time 30 min and using balls of diameter 5 mm only.

3.2 Morphology of milled silicate product

FE-SEM images of silicate powder before/after milling process are shown in Figure 2. In Figure 2(a), the coarse glass powder before milling process was found to be polygonal in structure. After the milling process, change to particle size and shape of milled sample particles had occurred, as shown in Figure 2(b). The particle size had become slightly smaller with good dispersion, specifically the milling speed at 500 rpm with operating time of 30 min and ball diameter of 5 mm were optimized parameters for the milling process. As expected from the particle size analysis in section 3.1, the silicate particles tended to aggregate with the increase of milling time with high accumulation of thermal energy related to the induction of large particles. Therefore, particles size distribution, and powder homogeneity are highly influenced by rotation speed, milling time and ball diameter [13-15].

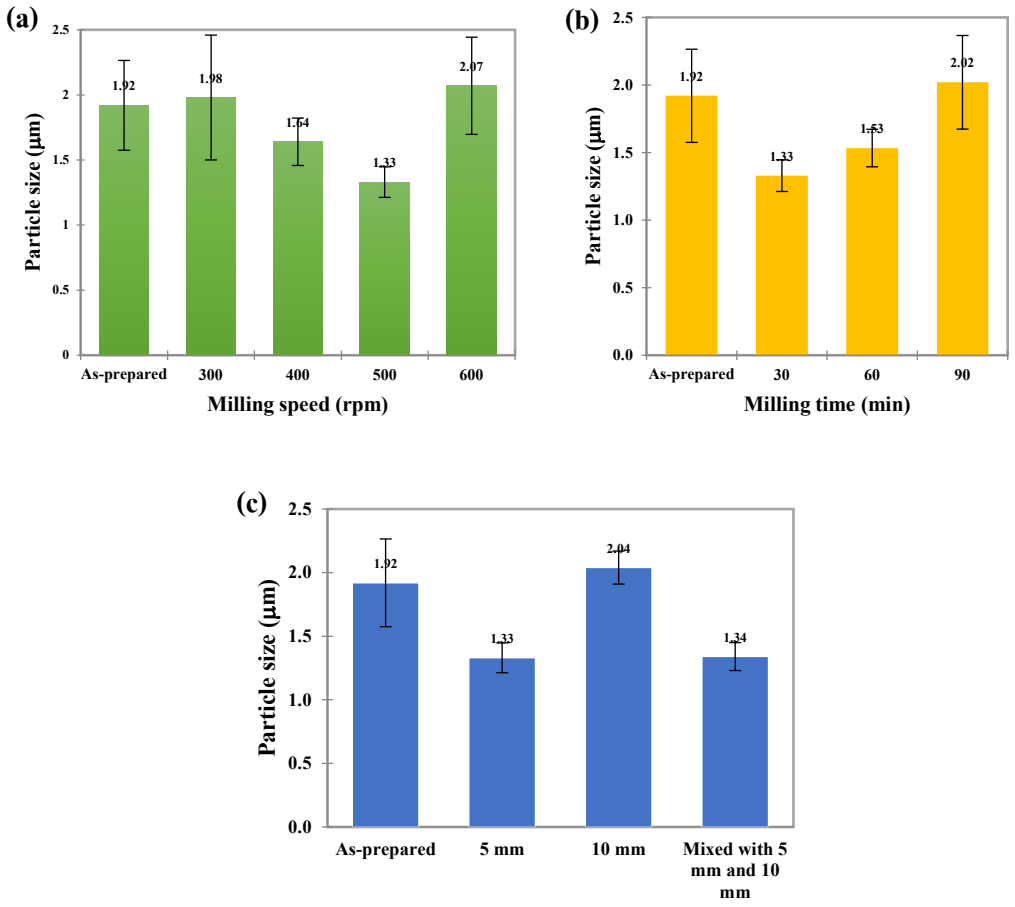

Figure 1. Average particle size of milled silicate powder with (a) different rotation speed at milling time of 30 min, (b) different milling time operating at 500 rpm and (c) different ball diameters operating at 500 rpm for 30 min

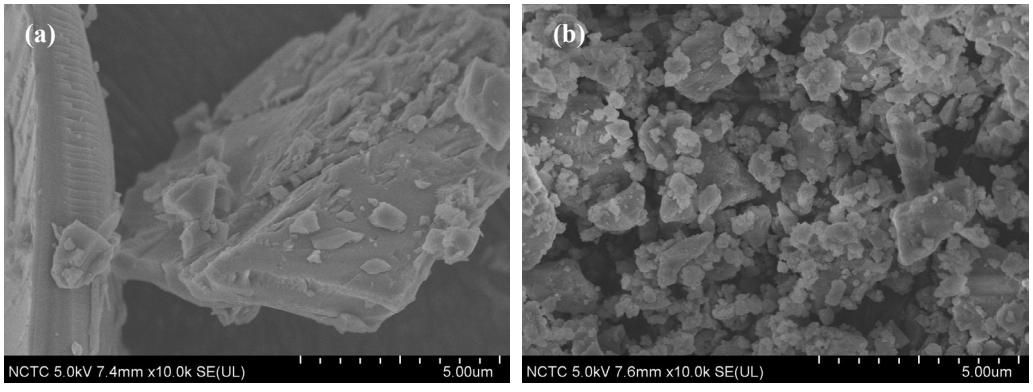

Figure 2. SEM images of silicate cullet (a) coarse powder before milling process and (b) milled product at rotation speed 500 rpm for 30 min with ball diameter of 5 mm

3.3 Functional groups of milled silicate powder

Functional chemical groups of milled silicate samples with different milling speed, time and ball size diameter were analyzed by FTIR spectroscopy, and the results are shown in Figure 3. In Figure 3(a-c), the chemical bond structure of the silicate powder showed the same patterns of functional groups. The bands appearing at 459 cm^{-1} and 800 cm^{-1} were identified as Si-O-Si bending vibration, and O-Si-O symmetric stretching vibration, respectively [16, 17], and these were the main groups present in the beaker components. Then, the band at 669 cm^{-1} was assigned to the bending vibration of bridging oxygen (B-O) between trigonal BO_3 groups. This indicated the secondary phase in beaker components. The absorption band at 925 cm^{-1} was assigned to the B-O vibration of BO_4 units and also associated with the stretching frequency of Si-O-B linkages [1]. Moreover, the characteristic of symmetric stretching relaxation of the B-O band of trigonal BO_3 units was detected at 1397 cm^{-1}. Also, the spectral band at approximately 1085 cm^{-1} was assigned to tri-, tetra-, pentaborate and diborate groups belonging to BO_3 and BO_4 groups along with asymmetric stretching of Si-O-Si bonds. Therefore, these results confirm that the mechanical ball milling process is a suitable process for the size reducing of glass powder. Meanwhile, the increase of high energy absorption at each band had obviously occurred in the milled samples due to the effect of particle size reduction compared with the sample before milling process. A relationship between particle size and spectral signal intensity in absorption mode of FTIR spectra was reported by Tubilla and Walker [18] who revealed that higher intensity and spectral reproducibility for FTIR sampling techniques were observed as the effects of creating smaller particles. Meanwhile, a high specific area of milled silicate powder was generated via the milling process due to the decrease of particle size. This mechanism can be related to the increase of silicon ion generation in silicate powders by the breaking of surface silanol (Si-OH) and siloxane (Si-O-Si) groups bonds [19]. Therefore, the enhancement of FTIR intensity occurred in the milled silicate product compared to raw glass cullet.

3.4 Light scattering of milled silicate powder

The light scattering property and transparency of milled silicate powder in polymer solution at different milling speeds was investigated by luminance meter, and the results are exhibited in Figure 4. The crucial parameter of milled silicate powder by milling rotation speed was chosen for light scattering property in Figure 4(a) owing to the wide range of different silicate particle sizes. Meanwhile, the optimized particle size of silicate powder can be proposed for light scattering material for light guide plate application. The highest luminance occurred at milling speed 500 rpm with particle size 1.33 μm and average scattering intensity at 275 lux. Meanwhile, the luminance intensities of large particle sizes of silicate powder at rotation speeds 300, 400 and 600 rpm were 266, 267 and 241 lux. In the case of coarse powder, the lowest luminance intensity occurred due to its large particle size of approximately 2 μm. The significant difference in luminance values of the samples milled at 300 rpm and 600 rpm with comparable size could be due to irregularity of the surface of the milled samples. For the samples milled at the low speed, there could be more irregularity and rougher surface than the sample milled at higher speed. The rougher surface of the scattering particles correspondingly leads to a greater diffuse reflectance rather than specular reflectance and consequently greater value of total luminance value. Therefore, the particle size of the scattering material is a specific factor corresponding to Mie scattering theory. This type of scattering occurs when the atmosphere is composed of spherical particles equal in diameter to the size of the transmitted radiation relating to a complex interference pattern of scattered wavelets [20].

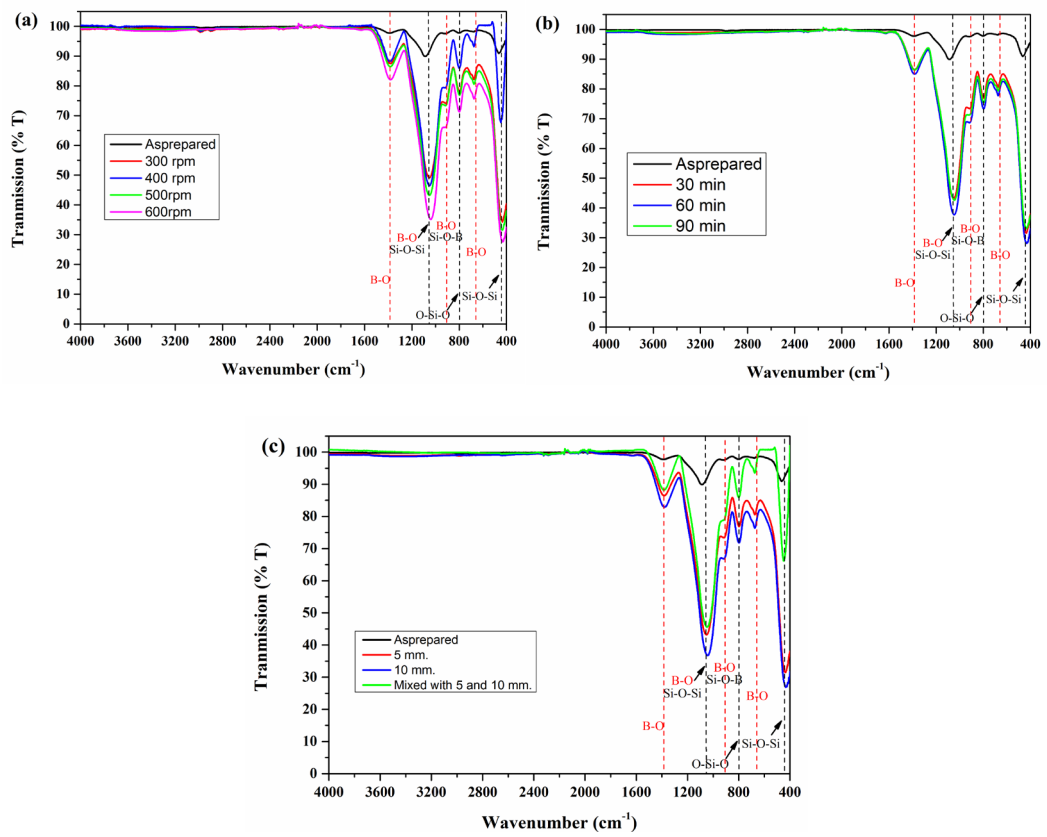

Figure 3. FTIR spectra of milled silicate powders (a) different rotation speed with milling time of 30 min, (b) different milling time operated at 500 rpm and (c) different ball diameters operated at 500 rpm for 30 min

However, the increase of luminance intensity can be obtained by further addition of scattering material in the matrix. Besides, transmission of silicate powder in solution with different milling speeds was observed by transmittance mode in a UV-Vis spectrophotometer as show in Figure 4(b). Under overall condition, high transparency of the solution had obviously occurred to around 98-99% level. This result indicated that good particle dispersion had taken place in the matrix. Meanwhile, the influence of optimized surfactant with suitable content in the matrix could help good dispersion of the scattering particles in the solution. However, the intensity of solution transmittance was decreased by the effect of a large amount of scattering material and additive polymer. Therefore, the optimized luminance property and transmission of scattering material depends on the light scattering application requirements. For high transparent matrix, a low content of scattering material is required. In case of high luminance in the matrix, a large amount of scattering material should be loaded, and this relates to more opacity in the matrix.

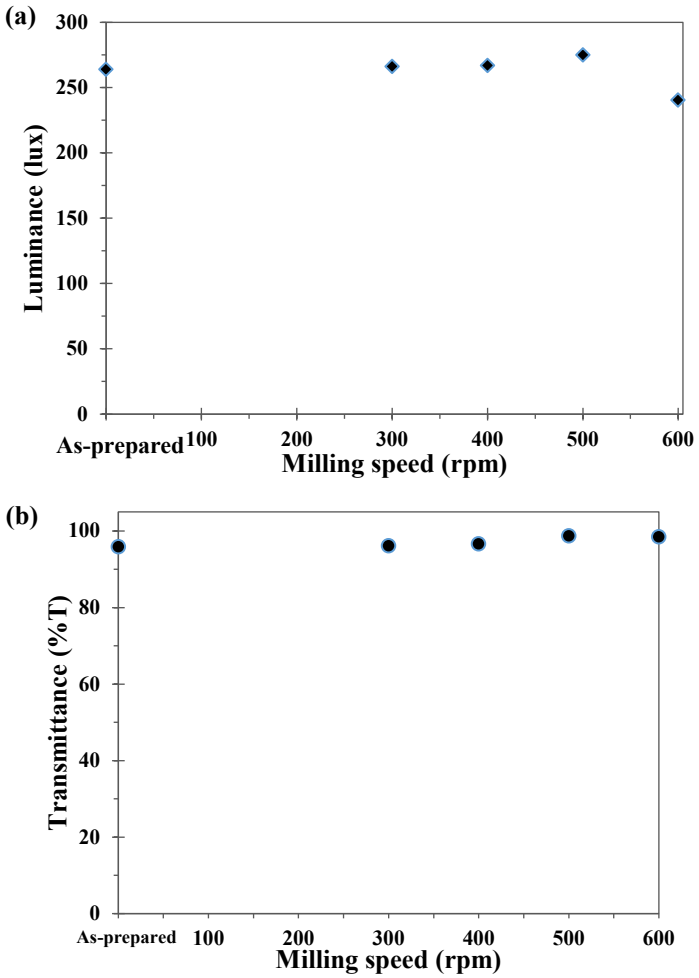

Figure 4. (a) Light scattering property and (b) Transparency of milled silicate powder with different milling speed in the solution

4. Conclusions

A fine powder in silicate product from waste beaker glass cullet was successfully prepared by a high energy ball milling process. The particle size of the milled silicate powder was reduced to 1.33 μm after milling at 500 rpm for 30 min with ball diameter of 5 mm. It has been observed that size reduction of glass cullet was highly dependent on the important milling parameters of rotation speed, time and ball diameter. While FE-SEM images indicated that the surface of silicate powder displayed fine particles that were much smaller than coarse powder (as-prepared sample). For FT-IR spectra, the functional chemical structures of milled powder showed the same patterns for all crucial milling parameters. This result confirmed that the chemical structure of silicate product had not changed with the application of mechanical force inder the high energy milling process. Moreover, the increase in height of absorption peaks in FTIR spectra observed was consistent with

the particle size reduction and high surface area of milled silicate powder. Furthermore, the scattering property was improved by milled the glass cullet at 500 rpm for 30 min, and the mill material showed high transmission at 99%, which corresponded well with the particle size results and SEM images.

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