Production of Fine Particles of Silica Glass and Limestone by Autogenous Grinding with A Stirred Mill.

—Effect of Mechanical Properties—

by

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ABSTRACT

The stirred mill is regarded as one of the most efficient devices for micronizing materials and has come to be actively used for preparation of fine particles. Recently, the demand for ultrafine particles is increasing in many kinds of industries.

In this paper, an autogenous grinding to get submicron or micronized particles based on frictional action of feed material has been carried out using a stirred mill. The mill used was made of stainless steel and its volume was about 5.5 liters and has 12 impellers. Samples used were silica glass and limestone. The feed mass was 5kg and the feed size was 20～13mm. In the present work, a operating parameter was stirring speed. The ground products were sieved in the size range from feed size to 325mesh (45µm), and from 45 to 0.17µm, the size distributions were measured by a laser diffraction and scattering method. The progress of grinding was evaluated by the increasing rate of fine particles. The effect of properties of samples on the grinding rate was studied.

As a result, it was found that an autogenous grinding with a stirred mill was more effective method for producing fine particles of limestone which is not elastic solid than silica glass which is nearly perfect elastic solid.

Key Words: Stirred Mill, Autogenous Grinding, Fine Particles, Silica Glass, Limestone

1. INTRODUCTION

The demand for fine or ultrafine particles keeps increasing in the fields of ceramics, composite materials, surface treatments, electronics, mineral processing and so on. The preparation of fine or ultrafine particles is accomplished by breaking-down or building-up. Breaking-down processes are very widely applicable, but it has been well known that the control of size and shape of produced particles are often difficult.
In general, the energy required for breaking-down, that is, the size reduction energy, the grinding resistance and the grinding rate, are expressed by a function of a particle size\(^1,2\). The energy efficiency decreases with a decrease in produced particle size. The development to find energy-saving size reduction processes or devices have been performed for years. One of these, stirred mills are now being used in mineral processing and coal industries\(^3, 4, 5\). In this technique, a large number of small grinding media are agitated by a pin-shaped impeller in a cylindrical vessel. Breakage occurs mainly by collision as captured between balls. On the other hand, submicron particles had been obtained easily by the mutual friction grinding of two specimens of the same materials\(^6-9\).

In a previous paper\(^10\), based on frictional action of feed material, a production of submicron particles by autogenous grinding with a stirred mill was investigated for limestone which is not elastic solid.

In this study, a autogenous grinding has been also investigated. The samples used were silica glass and limestone. The objective of this study has been to investigate the effect of mechanical properties of samples on the production of fine particles.

2. PROPERTIES OF SAMPLES USED

The samples used in this investigation were silica glass and limestone. Silica glass was selected as the nearly perfect elastic solid and limestone was not elastic solid\(^10\). Their properties are tabulated in Table 1. A production of fine particles by autogenous grinding with a stirred mill is based on frictional action of feed particles. Then, it could be considered that the mechanical properties have effect upon the production of fine particles.

Yashima et al\(^10\) had measured experimentally a conversion of fracture energy into plastic deformation energy by using specimens of sphere. The conversion percentage of silica glass and limestone are also tabulated in Table 1. Silica glass and limestone were classified manually by sieved to get the samples sized from 13 to 20mm.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Particle density (kg/m(^3))</th>
<th>Moh's hardness ( - )</th>
<th>Vicker's hardness (Kg/mm(^2))</th>
<th>Young's modulus (Pa)</th>
<th>Poisson's ratio ( - )</th>
<th>Compressive strength(^a) (Pa)</th>
<th>Compressive strength of sphere(^b) (Pa)</th>
<th>Shearing strength(^c) (Pa)</th>
<th>Work Index (kWh/ton)</th>
<th>Hardgrove Grindability Index ( - )</th>
<th>Percentage of plastic deformation energy ( - )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Silica glass</td>
<td>2.15 x 10(^3)</td>
<td>6-7</td>
<td>465</td>
<td>7.35 x 10(^{10})</td>
<td>0.16</td>
<td>5.15 x 10(^8)</td>
<td>2.70 x 10(^7)</td>
<td>&quot;</td>
<td>14.8</td>
<td>-</td>
<td>0</td>
</tr>
<tr>
<td>Limestone</td>
<td>2.70 x 10(^3)</td>
<td>4</td>
<td>115</td>
<td>6.80 x 10(^{10})</td>
<td>0.32</td>
<td>9.57 x 10(^7)</td>
<td>4.15 x 10(^6)</td>
<td>2.09 x 10(^7)</td>
<td>9.40</td>
<td>73</td>
<td>55</td>
</tr>
</tbody>
</table>

a) 2 x 3 cm  b) 2 x 4 cm  c) 2 x 5 cm

3. EXPERIMENTAL APPARATUS

Batch grinding experiments were conducted using an Attritor-Type D (Mitsui Mining Co., LTD) as shown in Fig. 1\(^10\). A torque meter was mounted on the drive shaft between the motor and the mill. Fig. 2 gives detail diagram of grinding apparatus.
The grinding conditions are tabulated in Table 2. The feed mass of silica glass and limestone were 5 kg. Samples occupied the bulk volume of which are about 95 % and 75 % of grinding vessel volume as shown in Fig. 2 with broken lines. The feed size was 13~20mm. This feed size had showed the minimum energy consumption to get submicron particles. The impeller speeds examined were 55, 69, 103, 139 and 173 rev•min⁻¹. The grinding times were two, four, ten, 25, 40, 60 and 180 minutes. The size distributions of each ground products were measured. Torque readings were recorded during each experiment, and the area under the torque vs grinding time curve was used to give a measure of the total energy input to the mill.

All products generated were sieved manually from feed size to four mesh (4.760 mm) and by a rotating and tapping shaker from six mesh (3.360 mm) to 100 mesh (0.149 mm). 10, 5, 5 and 3 g were sampled from the ground products finer than 100 mesh and were wet sieved manually with great care from 150 mesh (105 m) to 325 mesh (45 m), 4 sections. In a wet sieving, after drying at 45°C for seven
hours, the mass of the sieve with the particles was measured. In the size range from 44μm to 0.17μm, the size distributions were measured by a laser diffraction and scattering method.

5. RESULTS AND DISCUSSION

5-1. Torque-Grinding Time Curve

The change of torque of silica glass and limestone with grinding time are shown in Fig. 3, when the rotational speed of impeller was 55r.p.m. The torque of limestone is drastically changed with the grinding time as compared with silica glass.

5-2. Size Distribution of Ground Product

The variations of size distributions of ground products with the grinding time are shown in Fig. 4 in log-log scale, when the impeller speed was 55 r.p.m. The distributions of limestone show a wide distribution. Between 10^4 and 10 μm, the slope of distributions are relatively flat but less than approximately 10 μm the slope is 45°. In Fig. 4, it is recognized that the fine particles were directly produced by attrition, abrasion and so on from feed particles.

The slope of distribution of silica glass is different from that of limestone at the early stage of grinding but in latter stage, the size distributions of two samples are similar. The same results were obtained at all impeller speed.

5-3. Relationship between Mass Fraction of Submicron Particles and Total Number of Revolution.

Size reduction is an important step in many of the processes by raw materials are converted into intermediate or final products. Authors had discussed experimentally the energy efficiency to get fine particles by autogenous grinding with a stirred mill. In previous papers, the energy when the sample was not changed, was subtracted from the whole energy input to mill and the energy efficiency to get fine particles was discussed. The symbol "Tro" in Fig. 3 shows the torque when samples were not charged. In Fig. 3, "Tro" occupied the majority of the torque input to mill. It may arise from a consumption of torque to keep the stable revolution of impeller. The rate as well as the energy efficiency of grinding is important factor to evaluate the grindability of solid materials.
Fig. 4 Comparison of particle size distributions of silica glass and limestone and to control the grinding operation. Furthermore, it can be considered essentially that the energy input to mill was approximately proportional to the grinding time as shown in Fig. 3. In this study, first, the effect of impeller speed on the increasing rate of submicron particles was discussed experimentally when the feed mass and size of samples were constant.

The relationship between the mass fraction of submicron particles and total number of revolution is shown in Fig. 5. The increasing rate of submicron particles were independant of the impeller speed.
speed in both samples. The mass of submicron particles of limestone which is not elastic solid was more than silica glass which is elastic solid.

We applied the Tanaka’s equation\textsuperscript{15} which described the relationship between specific surface area produced and grinding energy or time, to the results in Fig. 5. Equations (1) and (2) were obtained.

\begin{align}
Q_i (G) &= \frac{W_i (G)}{W_s} = 0.0034 \left(1 - e^{-7.94 \times 10^{-5} n t}\right) \quad (1) \\
Q_i (L) &= \frac{W_i (L)}{W_s} = 0.077 \left(1 - e^{-9.85 \times 10^{-5} n t}\right) \quad (2)
\end{align}

where, \( Q_i (G) \) and \( Q_i (L) \) are the mass fraction of submicron particles of silica glass and limestone, and \( n \cdot t \), the total number of revolution. Equations (1) and (2) show that mass fraction of submicron particles approach gradually 0.0034 and 0.077 with an increase in the total number of revolution respectively.

5-4. Relationship between Mass Fraction of Fine Particles and Total Number of Revolution.

In autogenous grinding, it was recognized that the fine particles were directly produced from feed particles as shown in Fig. 4. In many kinds of industries, it can be presumed that fine particles are important materials as well as submicron particles for intermediate or final products. Then, we also investigated the relationship between the mass fraction of micronized silica glass and limestone and the total number of revolution.

Figure 6 shows the relationship between the mass fraction finer than 10\( \mu \)m and the total number of revolution. Equations (3) and (4) were obtained as well as the submicron particles.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig6.png}
\caption{Relationship between mass fraction finer than 10\( \mu \)m and total number of revolution.}
\end{figure}
where $W_{10} (G)$ and $W_{10} (L)$ are the mass of silica glass and limestone finer than $10 \mu m$.

The relationship between the mass fraction finer than $100 \mu m$ and the total number of revolution is also shown in Fig. 7. Equation (5) and (6) were obtained as well as the submicron particles.

$$Q_{100} (G) = \frac{W_{100} (G)}{W_S} = 0.174 \left(1 - e^{-1.38 \times 10^{-4} n \cdot t}\right)$$ (3)

$$Q_{100} (L) = \frac{W_{100} (L)}{W_S} = 0.353 \left(1 - e^{-2.50 \times 10^{-4} n \cdot t}\right)$$ (4)

Equations (5) and (6) show that the mass friction finer than $100 \mu m$ approach the same value with an increase in the total number of revolution. The values of coefficients in Eqs.(1)~(6) varied with the cut size. Then we assumed the following equation.

$$Q_X (S) = \frac{W_X (S)}{W_S} = a \left(1 - e^{\alpha n}\right)$$ (7)

where $W_X (S)$ are the mass of samples finer than size $X$. The relationship between $a$ or $\alpha$ and the cut size $X$ ($X = 1, 3, 5, 10, 30, 50$ and $100 \mu m$) were determined experimentally. Figures 8 and 9 show relationship between both respectively.

The mass friction finer than size $X$ was expressed experimentally as a function of $X$ and the
total number of revolution in both samples.

Fig. 8 Relationship between coefficient in Equation (7), \(a\), and cut size, \(X\), under grinding condition in Table 2.

Fig. 9 Relationship between coefficient in Equation (7), \(\alpha\), and cut size, \(X\), under grinding condition in Table 2.
6. CONCLUSION

In this study, an autogenous grinding to get fine particles of silica glass and limestone has been carried out using a stirred mill. The effect of mechanical properties of samples on the grinding rate was investigated when the feed mass and size were constant. The results are summarized as follows:

1. The mass of fine particles approached gradually the values which depend on the cut size and samples with an increase in the total number of revolution.
2. The increasing rate of mass of fine particles was independent of the revolution speed of impeller but depend on mechanical particles of samples.
3. The increasing rate of mass finer than 100 μm was independent of samples.
4. The experimental equations which expressed relationship between the mass fraction finer than size X (X = 1, 3, 5, 10, 30, 50 and 100 μm) and the total number of revolution were obtained.
5. It was found that an autogenous grinding with a stirred mill is more effective method for producing fine particles which are not elastic than nearly perfect elastic solids.

Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
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<tbody>
<tr>
<td>a, α</td>
<td>coefficients in Equation (7) (-)</td>
</tr>
<tr>
<td>n</td>
<td>rotational speed of impeller (rev min⁻¹)</td>
</tr>
<tr>
<td>n•t</td>
<td>total number of revolution (-)</td>
</tr>
<tr>
<td>Q</td>
<td>mass fraction of under size (-)</td>
</tr>
<tr>
<td>Q₁(G), Q₁(L)</td>
<td>mass fraction of submicron particles of silica glass and limestone (-)</td>
</tr>
<tr>
<td>Q₁₀(G), Q₁₀(L)</td>
<td>mass fraction finer than 10 μm of silica glass and limestone (-)</td>
</tr>
<tr>
<td>Q₁₀₀(G), Q₁₀₀(L)</td>
<td>mass fraction finer than 100 μm of silica glass and limestone (-)</td>
</tr>
<tr>
<td>Qₓ(S)</td>
<td>mass fraction finer than size X (-)</td>
</tr>
<tr>
<td>t</td>
<td>grinding time (min)</td>
</tr>
<tr>
<td>Tₐ</td>
<td>torque (N•m)</td>
</tr>
<tr>
<td>Tₐₒ</td>
<td>torque when materials was not charged (N•m)</td>
</tr>
<tr>
<td>X</td>
<td>particle size (μm)</td>
</tr>
<tr>
<td>W₁(G), W₁(L)</td>
<td>mass of submicron particles of silica glass and limestone (kg)</td>
</tr>
<tr>
<td>W₁₀(G), W₁₀(L)</td>
<td>mass finer than 10 μm of silica glass and limestone (kg)</td>
</tr>
<tr>
<td>W₁₀₀(G), W₁₀₀(L)</td>
<td>mass finer than 100 μm of silica glass and limestone (kg)</td>
</tr>
<tr>
<td>Wₛ</td>
<td>feed mass (kg)</td>
</tr>
<tr>
<td>Wₓ(S)</td>
<td>mass finer than size X (kg)</td>
</tr>
</tbody>
</table>

References:

2) Bond, F.C.: Trans. A. I. M. E., 193, 484 (1952)
9) Morohashi, S., and S. Sawahata: ibid, 28, 804 (1979)