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Grinding of calcite suspensions in a stirred media mill: Effect of operational parameters on the product quality and the specific energy

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ABSTRACT

This paper investigates the production of calcite suspensions by a wet grinding process in a stirred media mill. The experimental set-up allows the circulation mode process in the presence of sodium polyacrylate as additive. The influence of different operational parameters on grinding results in terms of particle size distribution and rheological behavior of the suspensions as well as the grinding efficiency is presented. We observe that the specific energy input that leads to a required product quality is not the same. Moreover, analyzing the trend of particle size versus the specific energy input instead of versus the grinding time, different and opposite conclusions flow from the experiment results. In addition, the effect of particle mean size on the suspension viscosity is presented.

1. Introduction

In the recent years, the industrial demand for ultrafine particles increased a lot due to the specific properties of nanoparticles, particularly in the chemical and pharmaceutical industries where products with high homogeneity or solubility are often required. Among product properties, particle fineness, expressed by the median size or the width of the particle size distribution is of prime importance. But other properties, as the suspension stability or rheological behavior are also important parameters for industrial applications. Stirred media milling has proven its ability to produce ultrafine particles in concentrated suspensions from a coarser product [1–3]. In such mills, the fragmentation results from the compression and shear induced by high speed rotating grinding beads with an agitator. It is known that a lot of parameters can affect the results of wet grinding and dispersion in stirred media mills. These parameters can be classified into four groups: grinding chamber and stirrer geometries, operating parameters (grinding time, stirrer tip speed, grinding bead filling ratio, bead size and properties, …), grinding operation mode (continuous, batch, pendular or circulating mode) and suspension formulation (solid concentration, particle size, additives, …). Many papers deal with the influence of different operating parameters on the grinding results. Research is usually conducted with two main objectives.

First, work aims to analyze the performance of the mill [4–8]. This approach is very interesting to get quantitative information on the energy required to grind a product and it gives a very good basis for mill design and extrapolation to an industrial scale. Indeed, the grinding process mainly depends on the specific energy input (SE) which is the total energy supplied to the grinding chamber related to the mass of the ground product. Since wet grinding processes are extremely energy intensive, the optimization of the process parameters is important to minimize the energy consumption. For a given particle suspension, the quality and the fineness of the product that can be obtained by a milling process are determined by the number of stress events undergone by the particles of the suspension, known as stress number (SN) and the stress intensity at each stress event, also known as stress intensity (SI). Thus, the optimization of the process parameters for a desired particle fineness, in particular the grinding media materials, the bead size and the stirrer tip speed, is usually done by evaluating the specific energy or the stress intensity [6–9]. Moreover, as pointed out by Kwade [5], the particles to be ground through a grinding process and the resulting fragments are not subjected to the same number of stresses neither to the same stress intensities. Thus the number of stress events SN and stress intensity SI can only be characterized by distributions, depending on the operating parameters. The width of the distribution of stress number is determined above all by the residence time distribution of the particles in the mill. While the width of the distribution of the stress intensity depends mainly on how the stress energies differ locally and over the grinding time. Recently, Yamamoto et al. [10,11] used DEM simulation to determine the velocity distributions of beads in a stirred mill and correlate the size reduction kinetics with the bead impact energy.

Second, other research is focused on ground product quality (fineness, suspension stability, surface properties, …) in relation with the end-use properties of the products. This second approach involves more products with high added value as cosmetics or drugs, usually produced in small quantities. It is also of prime importance for the production of nanoparticles. Indeed, in the sub-micron particle

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size range the behavior of the product suspensions is more and more influenced by increasing particle–particle interactions. Due to these interactions, agglomeration phenomena can occur limiting further size reduction and a fixed minimum particle size is reached whatever the operating conditions [12]. For the same reasons, the viscosity of the ground suspension often increases [13] and this affects the ability of the particles to be broken. Also, a lot of work focuses on the research of grinding additives [14–16] or operating conditions able to improve suspension stability during the grinding process [17]. In other recent papers, a bead milling process was even used to enhance dispersion stability [18] or to improve other end-use properties [19,20]. When the focus is on the properties of the ground product, the analysis of the grinding kinetics regarding the evolution of the particle size distribution or other desired properties versus the grinding time is of prime importance. Another important feature in stirred media milling is related to the bead wear and the possible contamination of the ground product [21,1], but this point is out of the scope of our paper.

In our work, we did not want to favor one or the other objective (energetic performance or product quality). Instead, we wanted to connect the two types of analysis to better understand the influence of some operating parameters and assist in their selection. Calcium carbonate was chosen as a model material since its behavior during stirred media milling is well representative of the majority of inorganic or organic materials. It is obvious that many parameters related to the mill geometry, the operating mode and to the formulation of the suspension, affect the grinding result. In this study, we investigated the influence on the grinding process of some of these different operating parameters and both interpreted the results in terms of grinding kinetics and specific energy. The rheological behavior of the suspension during the grinding process of particles is also studied for different operating conditions. Finally, the effect of particle mean size on suspension viscosity is highlighted.

2. Material and experimental device

A high pure (>99%) calcite ($CaCO_3$) obtained from Merck, Germany was used for the experiments. The density of this material is 2656 kg/m^3 at 20 °C and the median particle size at the initial state is about 30 μm (measured by laser diffraction). Moreover, sodium polyacrylate (SPA) from Sigma-Aldrich with a molecular weight of 5100 g/mol and a density of 550 kg/m^3 was used to disperse the particle of $CaCO_3$ in water. Garcia et al. [14] have shown that the use of this polyelectrolyte could help in processing calcite suspension avoiding the re-aggregation of fine fragments. The SPA concentration was kept constant at 8% (i.e. 8 g of SPA per 100 g of $CaCO_3$) for all the runs.

All the grinding experiments were performed in a laboratory stirred media mill (Labstar from Netzsch) using the circulation mode (Fig. 1). The initial suspension is put into a feed tank equipped with an agitator to ensure a good mixing preventing the formation of deposits or dead zones. During the run, the suspension is continuously pumped from the feed tank trough the stirred media mill by means of a peristaltic pump. The outlet of the mill is also connected to the feed tank allowing the ground product to go back into the feed tank. The power input delivered by the motor (M), the pressure (P) and the temperature (T) at the inlet and the outlet of the grinding chamber were continuously registered during the process.

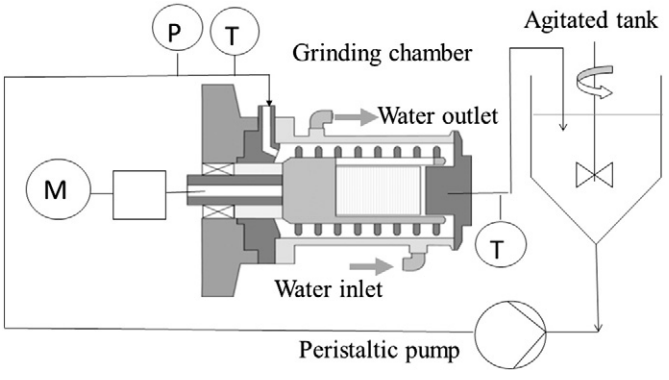

Fig. 1. Experimental set-up.

The determination of the specific energy depends among others on the mode of grinding operation. For a grinding process in a circulating mode (multi-passes), the specific energy can be determined as follows:

$$SE = \frac{\int_0^t (N(\tau) - N_0)\mathrm{d}\tau}{m_P} \quad (1)$$

Where m_P is the mass of the solid product, $N(\tau)$ is the power at the time τ and N_0 is the no-load power. If the grinding media wear is significant, especially, considering long grinding periods, a correction can be introduced [22] to take into account of it in the expression of the specific energy. In that case, it is assumed that the media wear increases proportionally to the power input N and the mass of the product is then extended by $0.5\Delta m_{GM}$, with Δm_{GM} the grinding media wear at the end time t of the grinding process. For our experimental study, the bead wear has been reduced, choosing high resistant beads (Y_2O_3-stabilized ZrO_2 media) and by using a grinding chamber and a separating cartridge lined with Cr–Ni-steel and equipping the agitator with tungsten–carbide. During the runs, the torque and the number of revolutions were automatically measured by a torque sensor put on the stirrer shaft, thus allowing calculating the specific energy using Eq. (1).

Moreover, samples were withdrawn over the run duration at the outlet of the grinding chamber. Particle size distribution was characterized by photon correlation spectroscopy (PCS) using a Nanosizer ZS (Malvern Instruments) in the nanometer size range. This apparatus gives access to the hydrodynamic diameter of the particles on the base of the Brownian diffusion.

Rheological measurements of the ground suspensions were also performed at ambient temperature using a rotational rheometer AR2000 (TA Instruments). A cone and plate geometry (60 mm diameter, 2° cone angle) was used. A pre-shearing step was imposed to ensure the same starting point for all the samples and then the shear rate was linearly decreased from 1000 to 0.1/s.

3. Effect of operational parameters

3.1. Effect of the volume flow rate

The effect of the volume flow rate on the grinding result is first discussed. Using the circulating mode, the residence time by pass and the number of passes are modified changing the flow rate. The residence time τ_p, of the suspension inside the grinding chamber by pass is equal to:

$$\tau_p = \frac{V_{GC} - V_{GB}}{Q_S} \quad (2)$$

where V_{GC} is the volume of the grinding chamber equipped with the agitator and the cartridge sieving, V_{GB} the true volume occupied by the beads (equal to the number of beads multiplied by the bead volume) and Q_s the suspension flow rate.

The suspension volume V_s and the full operation time t being fixed, an increase of the flow rate implies a decrease of the suspension residence time by pass, but also an increase of the number of passes during the full operation time. Thus, the true residence time of the suspension during a grinding run in the circulation mode is given by:

$$\tau_s = t\frac{(V_{GC} - V_{GB})}{V_s}. \quad (3)$$