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Dry ball mixing and deagglomeration of alumina and zirconia composite fine powders using a bimodal ball size distribution

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Abstract

Agglomerates lead to poor and unreproducible properties of ceramics. To ensure a reliable manufacturing process and final product properties, a batch needs to be well deagglomerated and also well mixed. This phenomenological study compares the application of a bimodal ball size distribution to monomodal ones of large or small balls in a tumbling mixer operated at 30 rpm. The ball sizes were adapted to the agglomerated particle sizes using Bond's ball sizing relation. Additionally, the diameters of the small and the large balls were adjusted according to Furnas' densest packing theory. The used filling ratio was 33 vol% and the batch of zirconia and alumina filled the interstices between the balls. The agglomerated batch had a $d_{50} = 1.2$ μm and a $d_{99} = 5.1$ μm. The small balls ($d_{50} = 2.25$ mm) caused a good macroscopic mixing degree for conditioning 10–20 min as investigated by specific surface area and true density. After initial deagglomeration, reagglomeration occurred for times ≥ 10 min. Conditioning with large balls ($d_{50} = 15$ mm) led to a comparatively large $d_{99} = 2.2$ μm but reagglomeration was suppressed for times ≥ 20 min. The microscopic mixing degree was also good for times ≥ 20 min as evaluated by scanning electron microscopy and X-ray diffraction. The bimodal ball size distribution led to a time-insensitive $d_{99} \approx 2.0$ μm between 10 and 40 min. Consequently, the robustness of the deagglomeration process increased according to the approach of Taguchi. Therefore, the simultaneous optimization of mixing and deagglomeration was simplified to the mixing optimization. An excellent mixing degree was achieved for times ≤ 10 min. Therefore, the conditioning result by the bimodal ball size distribution was superior to applying monosized balls.

Keywords: Tumbling mixer; Ball size distribution; Cohesive powder; Deagglomeration; Ball mixing

1. Introduction

Mixing of granular materials is of great concern in many industries such as ceramics, metallurgy, polymers, composites, pharmaceuticals, food processing, and agriculture [1]. A central problem in these industries is dry mixing and simultaneous deagglomeration of fine cohesive materials due to poor movability and natural agglomeration [2–4]. Fine particles have a large specific surface area as well as a high chemical reactivity and physical attraction. Therefore, they have possible benefits for contemporary and future products, which is why research on mixing technologies of fine particles is of predominant importance [3,5].

In ceramic technology agglomerates influence the rheology [6,7], limit the attainable density [8,9], lead to localized nonuniform shrinkage and therefore to poor mechanical properties [10–14]. Hence, agglomerates need to be eliminated to ensure a reliable manufacturing process and reproducible properties of ceramics [6,12,14].

To break down agglomerates and to mix fine particles, high energy and forces are applied by moving container or tools of the mixer [15,16]. For hydratable powders like alumina dry conditioning is preferred because it causes less microscopic defects after firing compared to wet processing [10,17].

Previous work in the field of dry conditioning has focused mainly on combined deagglomeration and comminution of fine powders. Ball milling is common for grinding fine powders because contamination by wear can be minimized by choosing the ball material according to the batch material [17]. For

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example, Le Caër et al. [18] used a planetary ball mill with monosized steel balls of 16 mm in diameter with a rotational speed of 640 rpm to batch metal powders. Krycer and Hersey [19] equipped a vibratory ball mill with nearly monosized alumina balls with a diameter of 23.2 ± 1.4 mm at 2800 rpm. Depending on the pursued size reduction and the process parameters the rotational speeds and conditioning times were adjusted.

More recently, compulsory mixers were studied for the preparation of nanoscale materials. Daumann et al. [20] built a mixer with 50 rpm container speed and up to 4000 rpm tool speed to prepare powders between 12 and 21 nm in less than 2 min.

However, milling of fine powders consumes more energy than pure deagglomeration. To our knowledge, pure dry deagglomeration has been addressed only by few published studies. For example, Ferkel and Hellmig [21] used a planetary ball mill with monosized alumina and zirconia balls of 10 mm diameter with a rotational speed of 150 rpm for 10 min to deagglomerate alumina and zirconia powders with median diameters of 14 and 13 nm. Although the conditioning times were different, the speed of 150 rpm for deagglomeration was low compared to the necessary speeds of up to 4000 rpm to mill powders to a comparable size.

From milling technology it is known that a steady state of fragmentation and reagglomeration is reached depending on the process parameters [19,20,22]. Reagglomeration can be retarded by different techniques. One is to reduce the ball size successively as the grinding proceeds into finer regions because for a maximum grinding efficiency the ratio of the size of the grinding medium to the particle size should be kept in certain limits [22,23]. For too large balls the grinding capacity suffers because the number of contact points decreases and the amount of extreme fines could increase. For too small balls the numerous contacts are too weak to break or deagglomerate the nipped particles [23].

Nevertheless, in the described studies only monosized balls of diameters between 10 and 25 mm were used. Consequently, with proceeding conditioning the ratio of ball to particle size changes and the process efficiency decreases. However, to our knowledge the use of a ball size distribution, providing different ball sizes for different stages of conditioning, has not yet been investigated.

The purpose of this study therefore is to investigate the influence of a bimodal ball size distribution adapted to the agglomerate size on the mixing and deagglomeration result of alumina and zirconia composite powders. Small balls, which initiate supposedly numerous microprocesses leading to an excellent dispersion, and large balls, inserting the necessary process energy for the deagglomeration, are applied to a tumbling mixer at three different conditioning times between 10 and 40 min. Therefore, the innovative combination of the bimodal ball size distribution enhances the mixing and deagglomeration result simultaneously. This result was compared to the ones when only utilizing monomodal ball size distributions of small or large balls. The powder composition used in this study to compare the results was one for an alumina-toughened zirconia with 15 wt% alumina and 85 wt% zirconia.

2. Experimental

Firstly, the raw materials alumina and zirconia and the batch were analyzed before conditioning. Afterwards the process parameters as revolution speed, filling degree and ball material were chosen. Based on the characterization of the raw materials the small and large ball sizes and amounts as well as the amounts of powder to add were calculated. After mixing, the batch properties were measured to compare them in relation to the values before conditioning.

The raw materials were alumina (Martoxid MR70, Martinswerk GmbH, Germany) and monoclinic zirconia (CS02, SEPR Saint-Gobain ZirPro, France) because both materials tend to agglomerate during storage. To determine the extent of the altering, measured particle size distributions and specific surface areas were compared to manufacturers' data (Table 1). Therefore, the specific surface area (SSA) was determined by the Brunauer–Emmet–Teller (BET) adsorption method (Area meter II, Stroehlein Instruments, Kaarst, Germany) according to the standard DIN 66132. The particle size distribution was measured by laser granulometry (Coulter LS 230, Beckmann-Coulter, Krefeld, Germany) according to the standard DIN EN 725-5. To minimize the effect of further altering powder properties, all ball mixing experiments were carried out with material from the same batches during a short period of 3 days. Additionally, scanning electron microscopy (SEM) (ESEM XL30FEG, FEI company, Netherlands) images were analysed for a clear determination of primary particles and agglomerates according to Wei et al. [3].

The batch composition of 15 wt% alumina and 85 wt% zirconia was a model composition to test the effects of the different ball size distributions. The model composition was adapted from alumina-toughened zirconia composites [24–26] due to the high Z-contrast of alumina and zirconia in backscatter electron microscopy images [27] and to allow a clear determination, no process additives were added.

After conditioning, only the mix can be measured. To evaluate the influence of the different ball size distributions on the mixing and deagglomeration result, the powder properties had to be compared before and after conditioning. Consequently, the batch properties before mixing were calculated from the measured data of the raw materials.

To evaluate the deagglomeration effect, particle size measurements and SEM investigations were performed. Agglomeration

Table 1
Raw material properties according to manufacturers.

Property	Alumina MR70	Zirconia CS02
d_{10}	0.1–0.4 μm	0.3 μm
d_{50}	0.5–0.8 μm	0.8 μm
d_{90}	1.5–3.0 μm	1.6 μm
SSA (BET method)	6–10 $m^2\ g^{-1}$	5 $m^2\ g^{-1}$