IFPRI
International Fine Particle Research Institute

Technische Universität Braunschweig

A Systems Engineering Approach to Dry-Milling with Grinding Aid Additives

Renewal Project Proposal

2022

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1. Introduction

This proposal is a continuation of the ongoing IFPRI research project on a system engineering approach to dry grinding, that aims to develop methodologies for understanding, optimizing and scaling such processes. In terms of process units, the current project phase focus on a dry grinding circuit based ball mill and air classifier. A special attention is paid on the manipulation of the material flow and cohesion properties by addition of surfactants, here called grinding aids. Thus, this project does not only cover traditional process aspects such as mill and plant design and simulation, material resistance to breakage and operation parameters, but also powder flow, cohesion, as well as energy flows and dissipation within the milling and classification units.

The project work for the initial three years was divided in three main work packages:

a. Identify which aspects of the ball milling process are affected by different GA

b. Identify which aspects of the air classification process are affected by different GA

c. Modify process models from the literature to account for the presence of GA, implement a flowsheet simulation tool and validate the flowsheet with mill-classifier circuit data

2. Summary of the current IFPRI project (2019-2023)

The powder feed materials selected within this project were calcinated alumina from Almatis Inc. and a calcium carbonate from IMERYS Minéraux Belgique SA. The selection of these materials in cooperation with IFPRI members were based on the differences in material hardness and bulk properties, such as powder flowability, with the alumina presenting higher hardness and better flow behavior in particle size distribution supplied.

Grinding aids (GA) are defined as liquid or dry substances that are added to the powder in order to increase the product throughput, decrease the specific energy consumption and/or to allow a certain product fineness to be reached. Three organic compounds in liquid form were selected as grinding aids from different substance classes, as presented in table 1. Although commercial grinding aids are in general provided as a blend of substances, within this project the additives are investigated in their pure form, since the goal is not to determine the best grinding aid available, but to establish a characterization and modelling procedure based on known chemistry of the grinding aid substances. The selection of this particular substances were based on previous studies from the literature by taking into account their distinct contribution on grinding product fineness, powder flowability and powder covering of grinding media after milling [1].

Table 1 – Grinding aids used in the project

Substance class	Compound	Purity [%]	Boiling point [°C]
Glycol	Diethylene glycol (DEG)	>99.0	244
Carboxylic acid	*n*-Heptanoic acid (Hepac)	>96.0	223
Alcohol	1-Hexanol (HexOH)	>98.0	157

2.1. Impact of grinding aids on milling

For the grinding experiments two lab-scale tumbling ball mills were selected, a 4 L batch mill and a 47 L continuously operated mill. The ball charge for all tests were alumina balls. Figure 4 compares the grinding efficiency between the pure material and the materials mixed with additives during batch milling. The advantage of the batch milling trials is that material transport can be ignored, so that the direct impacts on grinding mechanisms of powder flowability and reduction of caking on the mill shell are more evident.

It is evident that some classes of GA are more effective in reducing energy consumption to achieve a given product size. In the case of alumina, the alcohol and glycol classes promoted higher grinding efficiency, while the carboxylic acid resulted in negligible efficiency improvement compared to grinding without GA. For the calcium carbonate, the all additives result in similar effects on grinding, still improving efficiency in comparison to no additive. However, it must be pointed out that the calcium carbonate had a feed size (x50 ≈ 10 µm) closer to the size limit which is achievable by this laboratory ball mill.

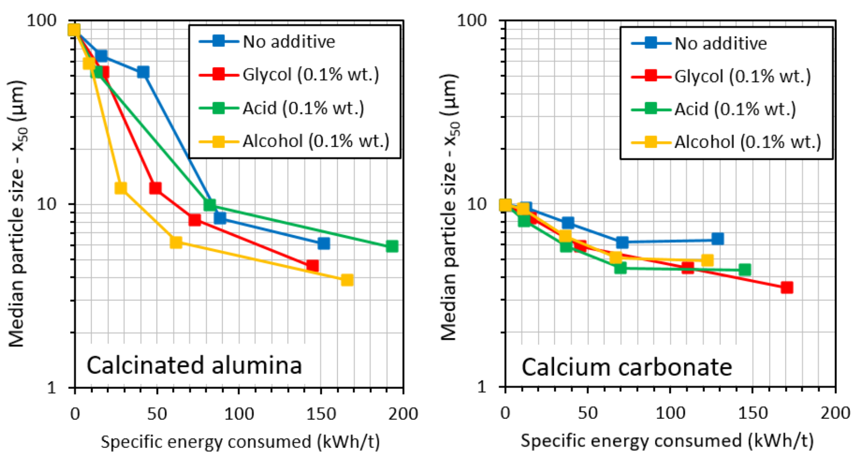


Figure 1 – Impact of additive type on the batch grinding results at 70% of critical speed, 30% media filling and 100% media void filling by product.

The products of batch milling trails were used for powder flowability measurement, conducted using a Schulze ring shear tester. Figure 2 compares the flowability index (ffc) for the samples with the different additives. It is interesting to observe that, although the carboxylic acid promoted a notable increase in flowability for both materials, in the case of alumina grinding, only a small effect in terms of milling efficiency was observed, whereas for the calcium carbonate this substance resulted in an increase of efficiency.

Prziwara et al. (2020) verified experimentally that the powder flowability index has an inverse relation to the amount of material stressed between two colliding balls. In other words: a better flowable powder can more easily escape out of the gap between balls and result in reduction of grinding kinetics.

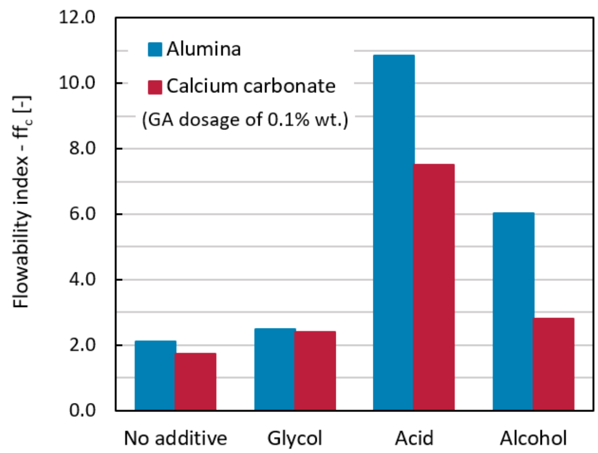


Figure 2 - Flowability index of grinded product in relation to additive type, measured with a Schulze ring shear tester with pre-consolidation of 1.0 kPa.

In the case of continuous ball milling in open circuit, the impact of the grinding aids carboxylic acid (high flowability) and glycol (low flowability) were compared to process performance without GAs. Figure 3 presents the process dynamics in terms of evolution of throughput and product particle size. For alumina, when no GA was used, caking of material on mill walls was intensive enough to result in process instability and clogging of the discharge outlet. With the use of carboxylic acid, the opposite occurred. Although stability was achieved fairly quickly, virtually no grinding was observed, indicating that the powder traversed the mill length so rapidly that there was not enough residence time for meaningful grinding. This fact was also underlined by the very low mill holdup measured. Interestingly enough, the glycol type of GA resulted in the best mill performance among the tested cases, in terms of product size.

When calcium carbonate was ground, the three tested cases showed similar results, with similar product sizes. Nevertheless, a reduction in process stabilization time was observed with the use of GAs. Considering that the feed median particle size was in a range in which high powder cohesion is typically observed for this material, the final powder flowability was not high enough to compromise grinding when the carboxylic acid was used.

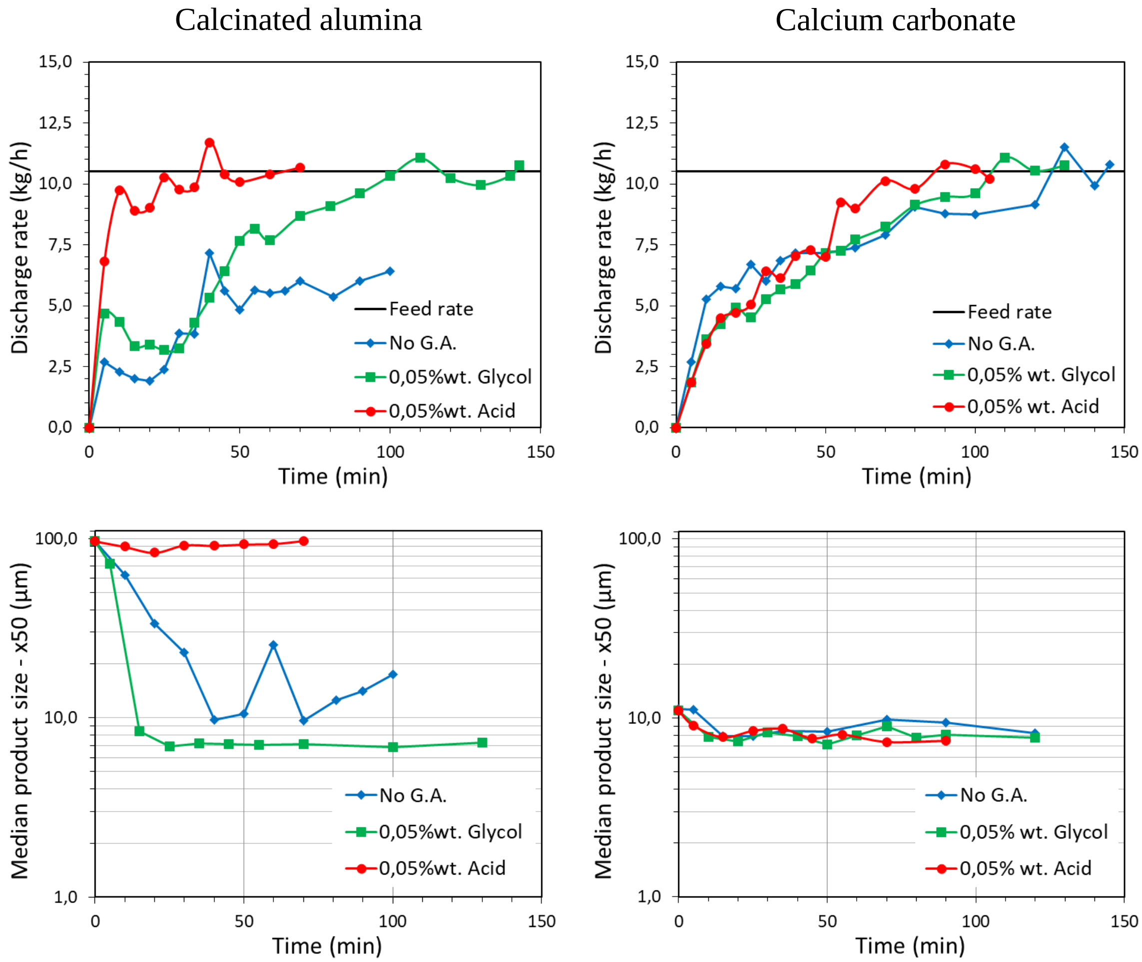


Figure 3 – Open circuit ball milling results.

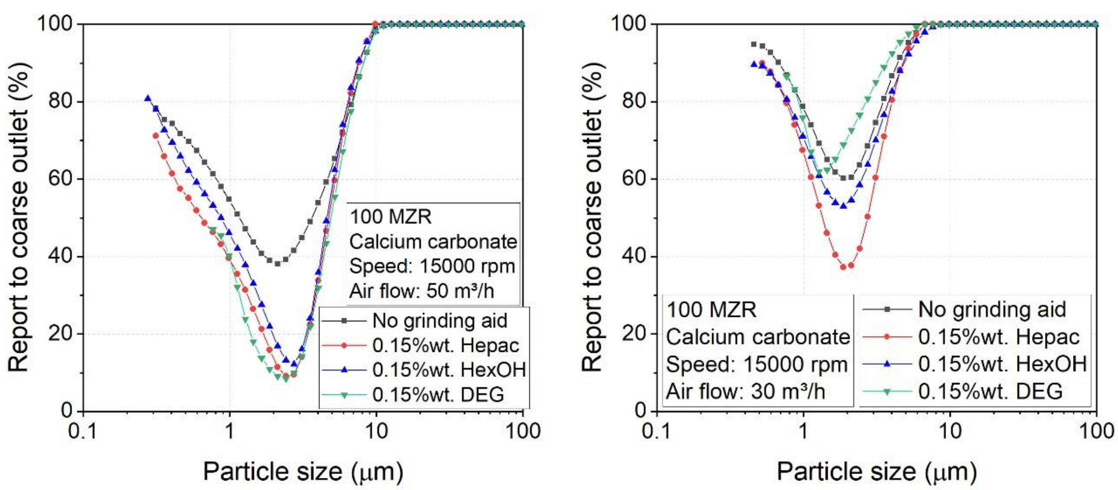
Although not all results observed in this work package are included in this proposal, its main outcomes can be summarized as:

- Powder flowability should be kept within an intermediary range (easy-flowing). However, both excessive and too low flowability should be avoided in order to improve throughout and process stability.
- High flowability is also detrimental to stressed material amounts and energy transfer from ball to product particles.
- Beyond flowability, GAs should be selected in order to reduce caking on equipment surfaces and reducing ball coating. Once an amount of powder is stressed between two ball, it should be readily freed to allow another sample of powder to be stressed.
- Grinding aids should be dosed in accordance with powder surface area, with surface coverage above 1.0 $N_A/m^2 \times 10^{18}$ presenting good results.

2.2. Impact of grinding aids on air classification

The classification experiments were conducted in two rotating wheel based air classifiers in different size scales, a laboratory and an industrial scale. At this stage of the project, all classification experiments were conducted in passage-mode – without milling as in a circuit – in the intention to characterize the effect of the equipment design and grinding aids on the classification process aspects. The laboratory scale classifier was a Hosokawa Alpine 100 MZR and for the industrial scale trials, a Hosokawa Alpine Turboplex ATP 100 was used. Both equipment possesses the same classifying wheel diameter (100 mm), but differ mainly in airflow range and dispersion chamber volume, with the ATP 100 presenting higher air volume flows and a much bigger dispersion chamber. It must be clarified that the size of the chamber influences the residence time inside the device and for how long particle agglomerates are subject to turbulent air streams.

The classification results were analyzed through the classifier partition curves, also called tromp curves, and some of its characteristic parameters, namely bypass of fine particles to the course outlet, the corrected cut size and fish-hook effect. Figure 4 presents the tromp curves for two air volume flows in the laboratory scale. The curves are close to the maximum nominal wheel speed. Considering that the air volume flow is directly proportional to the cut size, in the case of 50 m³/h, all GAs reduced the bypass in comparison to pure material classification, with similar tromp curves. On the other hand, for smaller air flows, with corrected cut sizes around 3 µm, all formulations presented considerable increase in the bypass. However, the carboxylic acid showed the smallest increase. Such process conditions, close to the limit of the operating conditions, will be inefficient, independent of the usage of GA.



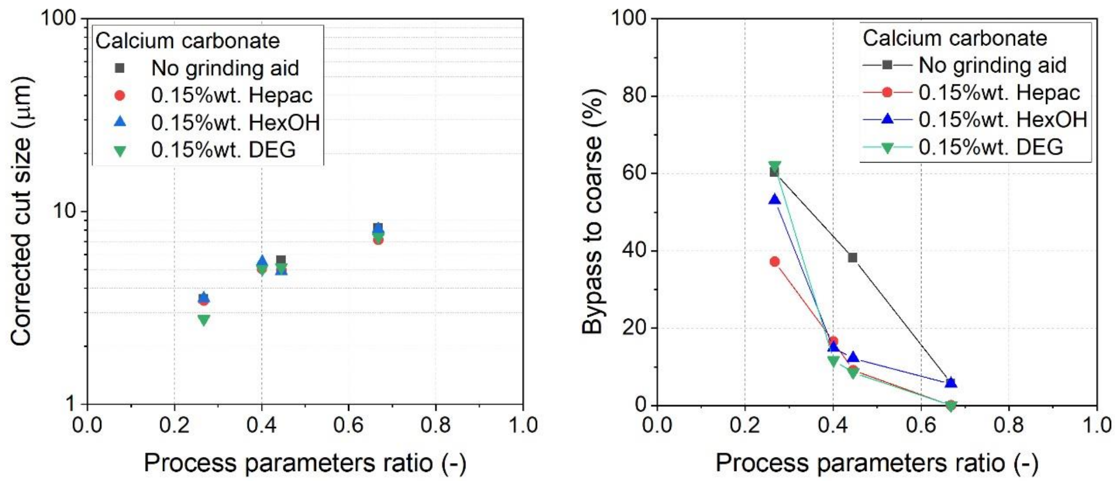


Figure 4 – Laboratory scale classification results. The two upper graphics are the tromp curves for two air volume flows. Below are the corrected cut size and bypass in relation process parameters.

In order to compare different classification conditions, the cut size and bypass were plotted against a dimensionless process parameter ratio (s. Fig. 4), which is calculated based on the equation below. A higher value of this ratio correlates with higher cut sizes and easier separation of the two products.

$$\begin{matrix} Process \\ parameter\ ratio \end{matrix} = \frac{air\ volume\ flow}{wheel\ area\ \times tip\ speed}$$

In Figure 4, the corrected cut size (when the tromp curve is compensated by removing the bypass) is not affected by the inclusion of GA to the powder. This finding is somewhat expected, since the main contribution of GA is the reduction of agglomeration and dispersion of fine particles. Therefore, the bypass is clearly reduced by GA, since this phenomenon is associated to not dispersed agglomerated particles. For high wheel speeds and low air flows, it becomes so difficult for particles to pass the rotating wheel, that most material report to the coarse outlet, no matter of the presence of GA.

In the case of the industrial scale trials, the intensity of powder caking was qualitatively investigated at the chamber internal surfaces. Figure 5 presents pictures taken from classifier chamber after process crash stop for two different wheel speeds. First, the caking intensity in the walls increased with increasing wheel speeds. Since high wheel speeds decrease the ability of particles in a given size to pass the rotating wheel, the residence time of finer particles inside the classifier chamber increases and also the likelihood of attaching itself to the walls. Second, the glycol did not contributed to meaningful caking reduction. This fact seems to be due to the chamber design and formation of air stream dead zones, but more studies are required to fully understand the effect. In contrast to the milling process, during operation material sticking on surfaces inside the classifier cannot be removed by contact with other media.

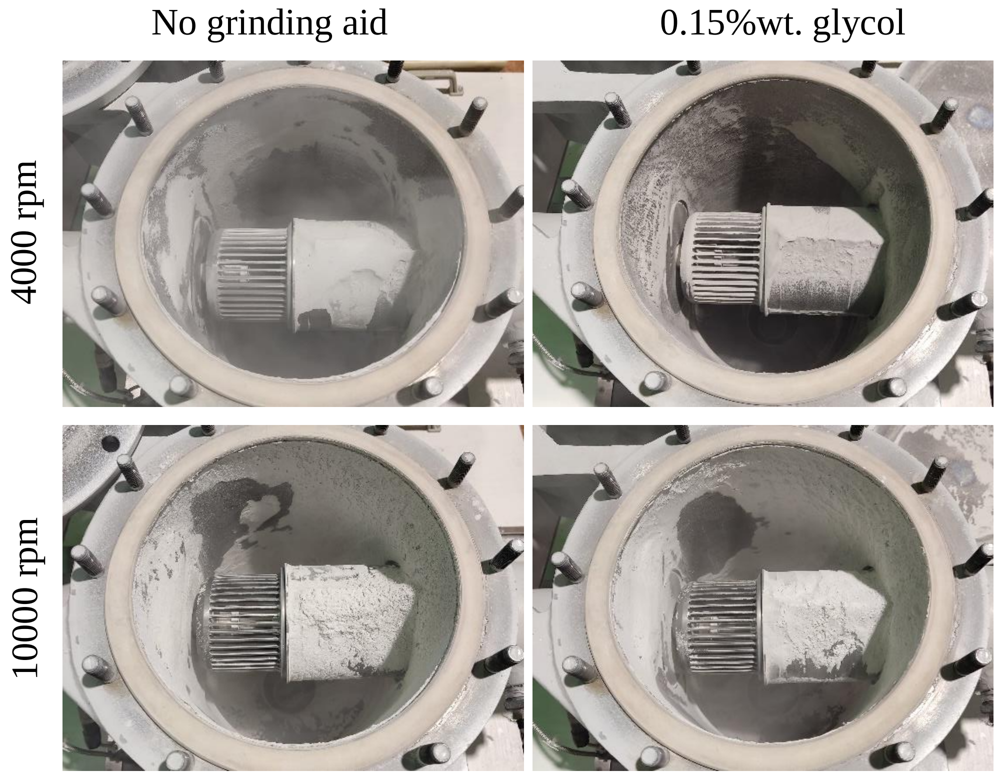


Figure 5 – Comparison of powder caking on classifiers chamber without (left) and with (right) grinding aid for two wheel speeds.

Figure 6 compares the partition curves at the industrial classifier for wheel speeds of 7000 and 10000 rpm. For the lower wheel speed, the curves bypasses were not significantly affected by the use of grinding aids. However, it must be point out that the bypass was small in both cases, indicating that better equipment design might be the one responsible for this results. For higher speeds, in a much more demanding situation for the equipment, the usage of grinding aids promoted a reduction of about 50% in the bypass.

Another aspect that can be observed is the separation sharpness for coarser particles, in the region between 20 - 40 µm. When GA was used, much sharper separation was observed, especially for 7000 rpm. This fact was not observed in laboratory scale classifiers, indicating that it was due to a combination between equipment design and grinding aids.

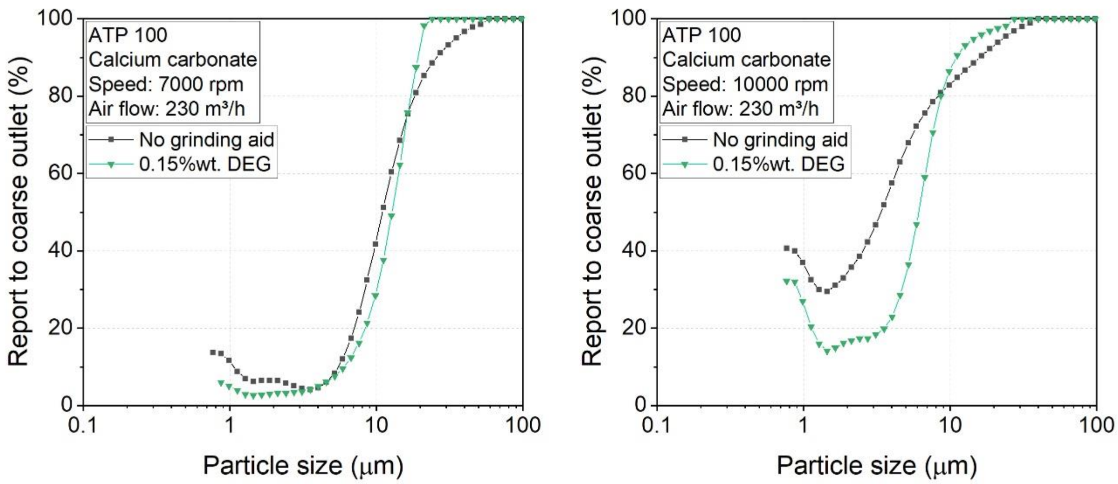


Figure 6 – Industrial scale classification results.

One last aspect to be compared between the two equipment is the fish-hook effect on the partition curves. This phenomenon can be observed by the increase in the curve for ultra-fine sizes (below 1 μm) on the left side from the bypass point. This increase of ultra-fine material reporting to the coarse outlet can be attributed to the formation of agglomerates, such as fine particles which are attached to the surface of coarser particles due to the surface charges. Looking at Figure 4 and 6, the laboratory scale classifier presented a higher fish-hook effect compared to the bigger scale classifier, even if GA is applied. Fine agglomerated particles require a minimum time of exposure to the internal air streams on the classifier to be dispersed properly. However, the small scale device consists of a very small chamber, so that the residence times are much shorter. Based on this observation, it can be concluded that the fish-hook effect is impacted much more by the classifier design then to the presence of GA.

The main outcomes of this this work package can be summarized as:

- The classifier corrected **cut-size is not affected by the use of grinding aids**, being a result of wheel speed and air volume flow rate;
- **Grinding aids promoted a reduction in bypass** of fines to the coarse outlet for both classifier scales, although with smaller intensity at very small cut-sizes;
- In the industrial scale classifier, **a reduction on powder caking on the chamber walls with grinding aids was not observed**;
- **The fish-hook effect for ultra-fine particles appears to be much more affected by equipment design**. Although, GA can improve it slightly;
- **Grinding aids promoted an increase in separation sharpness for coarser size classes**, reducing coarser residues on the fine product.

2.3. Milling and classification processes modelling with grinding aids

This work package is ongoing and will be completed by the end of the project in the Summer of 2023. In the following, the process models are presented which are developed and implemented in flowsheet environment. It is also presented the selected flowsheet software. Mill-classifier circuit trials are being conducted at the moment for future validation of flowsheet simulations.

In order to establish a flexible and reliable flowsheet tool, capable of predicting and allow optimization of the different ball mills in the scope of IFPRI, the models need to account for the process variables mentioned below:

- Mill geometry (diameter and length) and liners profile
- Mill speed and ball filling
- Balls material and size distribution
- Product material properties – particle strength and fragmentation
- Powder bulk properties – flowability and bulk density

The following requirements were defined for the flowsheet tool:

- Must predict product particle size distribution as a function of the process variables above
- Must allow simulation of a plant and equipment not yet in operation or in design stages

For the ball mill modelling, process variables such as mill geometry, liners profile, speed, ball filling and size has to be first translated into a dissipated stress distribution curve. This is done by means of Discrete Element Method (DEM) simulations of the mill. From the DEM simulation of the mill in steady-state, a stress distribution is extracted.

2.3.1. Ball mill model

The proposed mill model is formulated as mechanistic population balance for media mills capable of predicting grinding of particles sizes from the lower millimeter size range down to the sub-micrometer scale. The mechanistic approach to media mill models is a very flexible and powerful tool once it accounts for the contributions of the different stress and process mechanisms, allowing full separation of material and process aspects [3]. Based on experimental evidences, the following hypothesis were assumed as valid for this application:

a. The population of particles have a size dependent distribution of fracture energies – in other words each particle with a given size requires a certain value of applied energy to suffer fracture [3]
b. Particles are not stressed individually, but as part of a bed of particles captured between either two grinding media or a grinding medium and a mill surface [1]
c. All kinetic energy dissipated during a grinding media collision is transferred to the captured particle bed as applied stress [1].

Applied stresses from DEM simulation, product strength and bulk properties are accounted for in the breakage rate (S_i) over time for the fraction of material in size class i broken by applied stress energy E, integrated over all energies.

$$S_i(t) = \int_0^\infty \frac{m_{capt}}{m_{holdup}} \cdot N_{col}(E,t) \cdot \left[\int_0^1 F_i(eE) \cdot p(e) \cdot de\right] \cdot dE \qquad (2)$$

where N_{col} is the frequency of collisions between grinding media with dissipated energy E and can be estimated directly from DEM simulations. $F_i(eE)$ represents the fraction of material in size class i that breaks due to applied stress energy eE and $p(e)$ is the stress energy partition inside the stressed particle bed. $F_i(eE)$ is solely a material aspect and reliable models for this fraction, as a function of size are present in the literature [3]. The mass of stressed particle bed (m_{capt}) and the kinetic energy dissipation into the bed from colliding grinding media are highly affected by powder flowability and particle size distribution [4]. For instance, higher powder flowability and smaller adhesion to grinding media surface allows a bigger amount of powder to flow out of the capturing zone during a collision, resulting in a smaller captured bed mass. Prziwara et al. proposed that m_{capt} can be calculated as a function of grinding media diameter (d_{GM}) and impact velocity (v_{imp}). For the case of a collision between two spheres, m_{capt} is given by

$$m_{capt} = {}^{\pi}/_{4} \cdot \left(d_{GM} \cdot \sin\alpha_{capt}\right)^2 \cdot \rho \cdot x_{max} \quad \text{and} \quad \sin\alpha_{capt} = a \cdot {v_{imp}}^{b}. \qquad (3)$$

The parameters a and b, together with the powder bulk density (ρ) and the maximum particle size in the bed (x_{max}) are a function of the bed particle size distribution and powder cohesion, therefore, powder stressing and dissipation evolves alongside product size.

2.3.2. Air classifier model

In contrast to the ball mill, there is no standard design and operation principle for the air classification step. Therefore, it was decided to follow a general approach in this process simulation by utilizing a model to describe the tromp curve of the device. The formulation proposed by Whiten [5], presented in equation 4. The parameters C and d_{50c} are the fraction subjected to real classification ($1 - bypass$) and the corrected cut size, respectively. The parameters α, β and β^* must be fitted from experimental data for the material and GA combination.

$$E_c = C \cdot \left[\frac{\left(1 + \beta \cdot \beta^* \cdot {}^{d}/_{d_{50c}}\right) \cdot (\exp(\alpha) - 1)}{exp\left(\alpha \cdot \beta^* \cdot {}^{d}/_{d_{50c}}\right) - \exp(\alpha) + 2}\right] \qquad (2)$$

2.3.3. Flowsheet simulation system

As flowsheet software, it was selected the system Dyssol (Dynamic simulation of solids processes) developed by the Institute of Solids Process Engineering and Particle Technology, at the Hamburg University of Technology [6]. The selection was based on the following criteria and features:

- Dyssol is an open-source software and free for use at commercial and academic applications

- Allows implementation of new and private process units’ models
- Dynamic simulation of complex processes
- Consideration of solid, liquid, gas phases and their mixtures

For this project, the models briefly mentioned above were recently implemented into Dyssol. Mill-classifier circuit experimental results will be used to validate the simulation. Additionally, an industrial plant circuit will also be simulated, however, without experimental data for validation.

3. Proposal for project second phase

3.1. Objectives of the current proposal

a. Determine a proper powder microscopic property to represent the combination of material and GA type and dosage
b. Relate measurable microscopic particle properties with powder bulk behavior
c. Design a characterization procedure to obtain the model parameters required for axial transport simulation
d. Model powder internal axial transport during milling
e. Validated all developed models and flowsheet simulation with industrial data

3.2. Proposed work packages

WP1 – From Micro to Macro

Data such as grinding aid substance and applied dosage are numerical values that are not easily used on a process mathematical model. Considering that all findings from this project should be incorporated into a flowsheet environment at some point, the combination of grinding aids and powder material with a given particle size distribution must be represented by a property measurable in laboratory. Particle size distribution and surface area alone do not represent the presence of GA. Therefore, the best candidate so far is the specific surface energy [1]. At the Institute of particle technology (iPAT), this property is measured by Inverse Gas Chromatography (IGC).

This work package aims to establish a model to predict macroscopic bulk powder behavior from measurable particle properties such as, particle size distribution, specific surface area and specific surface energy.

In order to reach this goal, the first step is to determine which type of bulk solid experiment provides results that are affected by the three particle properties above and deliver useful information to predict flow during milling, i.e. between the approaching grinding balls and overall through the mill chamber. Since this project focuses on media milling, the bulk solids experiments would be conducted with mixtures of powder and grinding media in different proportions. The following devices for bulk solids experiment are available:

- Schulze ring shear tester
- Anton-Paar powder rheometer
- Granudrum – for dynamical angle of repose

- Granuheap – for heap angle
- Granupack – for dynamic tapped density analysis

Naturally, other suggestions of characterization tests proposed by IFPRI members, would be gladly considered.

The main outcome at the end of this work package would be the extension of the ball mill model to become able to also dynamically predict powder flow changes with evolution of product particle size. A possible first approach was proposed by Giraud *et al*. [7]. The authors presented a model that relates powder flow function coefficient (ff_c) with a population-dependent granular bond number (Figure 7a). This number is directly calculated from the powder particle size distribution and the Hamaker constant from the material. The latter is a function of surface energy. In Figure 7b, a dynamical change of ff_c with grinding time is presented.

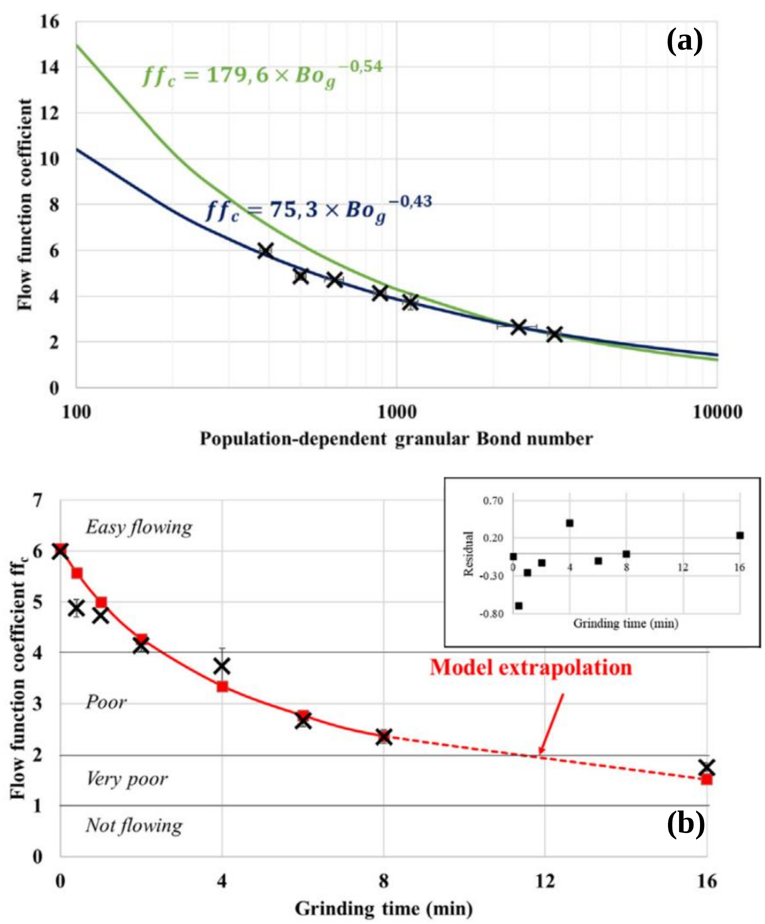


Figure 7 – Flow function coefficient of the alumina powder for different particles sizes, (a) as a function of the population-dependent granular bond number and (b) for different batch grinding times [7].

At the final stages of this work package, a standard characterization procedure will be proposed based on its findings in order to obtain the required material parameters for the process simulation, considering both, breakage and flow.

WP2 – Powder transport during milling and extension to a dynamical model

Currently, the mill model and flowsheet simulation tool requires the assumption of process steady-state. Specifically, the ball mill model depends on the product mass holdup and residence time as input parameters. Due to this limitation, initial tests on the mill to be simulated or previous data are required for flowsheet simulation, which might be critical for new product formulations or operations in designing stages. Considering that, this work package proposes the extension of the mill to a dynamical formulation.

To achieve this goal, the findings from WP1 will be used to formulate a powder axial transport model based on particle size distribution, powder flow behavior and process parameters (mill speed, mill filling and internal air volume flow). It will be also proposed a mill discharge function based on mill powder filling level, air volume flow and the mill outlet, more specifically, open area on the discharge grate.

Apart from bulk properties modelling, for the development of the transport and discharge model CFD-DEM coupled simulations of the ball mill in different operation conditions shall also be conducted to describe the impact of air flow on air swept mills.

Since the models require previous experiments on the ball mill only to describe transport with transport and discharge functions, the ball mill model and the flowsheet tool can be modified to a dynamical formulation, allowing process prediction and optimization, also for equipment still in design stage.

The mill model was formulated in a way that product, machine and operation contributions are completely separated. Thus, the flowsheet can be extended to simulate also dry stirred media mills. If that is in interest of the IFPRI members, additional effort would be done to account for the design differences between the two types of mills.

WP3 – Validation of flowsheet tool with industrial data

The experimental work done so far for this project was conducted in one pilot scale mill-classifier circuit. Although the plant at the iPAT allows control and measurement of almost all process variables, it is still a single plant in one size scale. Hence, this work package proposes the conduction of sampling campaign on an industrial grinding plant for later simulation and validation of the flowsheet tool.

This task can be coordinated with an IFPRI member that possesses a dry grinding circuit that, ideally, operates with GA. The work would be conducted by a small team from iPAT in collaboration with the IFPRI partner and following operations could be conducted:

- Collection of product samples in multiple points in the plant to measure particle size and mass flow rate;
- Measure of ball mill liner profile, including wear patterns, along the mill axis;
- Measurement of ball level and size distribution along the axis;

- Sampling of product along the mill;
- Sampling of product from both discharge of the classifier to obtain its efficiency curve;

Additional product should also be collected for material characterization in terms of breakage and bulk/flow properties following the procedure proposed in WP1. Lastly, DEM and flowsheet simulations of the plant is planned for models validation.

4. References

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