

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

## *IFPRI Research Project Proposal*

### **1. Introduction**

Many different aspects need to be considered for optimizing continuously operated dry grinding processes in terms of product quality, product throughput and energy efficiency. Thereby, the most important parameters can be found in a) the stressing conditions provided by the mill for a certain set of process parameters, b) the breakage behavior of the particles under the present stressing conditions, c) the particle and powder behavior during the stressing event, d) the material transport through the mill which influences aspects like the retention time distribution and the energy efficiency, as well as e) further process units within the grinding plant such as air classifiers and conveyors. Generally, the stressing behavior of a mill can be described by the kind of applied stresses (e.g. slow compression of single particles or particle beds, impacts ...), the frequency of stresses inside the mill as well as the stress energies. In detail, each mill can be characterized by a frequency distribution of the stress energies. In order to describe the effect of stresses per product particles, further aspects need to be considered, such as the number of stresses per product mass as well as the stress intensity (e.g. the area specific force or the mass specific energy acting in a single stressing event). Here, the mass of particles that is stressed in a single stressing event becomes very important. Moreover, the energy transfer inside the grinding machine has to be considered for any kind of dry-milling process: Only that proportion of the energy, which is transferred to the product particles and not dissipated into heat by friction or other aspects, can finally be used for stressing and breaking the particles.

Efficient stressing of particles is often complicated by attractive particle-particle forces which become more decisive with decreasing particle size. In general, these forces lead to a variety of challenges, in particular caused by a high state of agglomeration, a decrease of the powder flowability as well as material adhesions on the machine and plant equipment. It is well known that these aspects may complicate the comminution within industrial grinding plants on various levels simultaneously (e.g. regarding the grinding efficiency between the grinding tools, the performance of air classifiers as well as the material transport and retention time inside the single devices). In order to control these forces, so-called (mostly liquid) grinding aids, which reduce the particle-particle forces by adsorbing on the surface of the product particles, are often used as stabilizing additives. However, even though grinding aids are established in various industrial processes, their selection and application is still mostly based on empirical knowledge. A review article on the state-of-the-art of grinding aids, which is currently prepared by our institute on behalf of IFPRI, reveals that grinding aids may have strong impacts on several of the above-mentioned aspects. Especially the aspects listed under c) to e) may be strongly affected by the presence of those additives. As shown in the report, their mode of action is mainly caused by influencing various product properties, such as the powder flowability, the agglomeration tendency of the particles, the adhesion tendency of the particles on surfaces as well as their fluidization behavior. Thereby, various sub-processes inside the grinding plant may be strongly affected by these additives. Thus, an efficient selection of appropriate grinding aids is very complex and still not fully understood. This is further complicated by the fact, that significantly different grinding aid effects have been reported depending on the used milling type or operation mode.

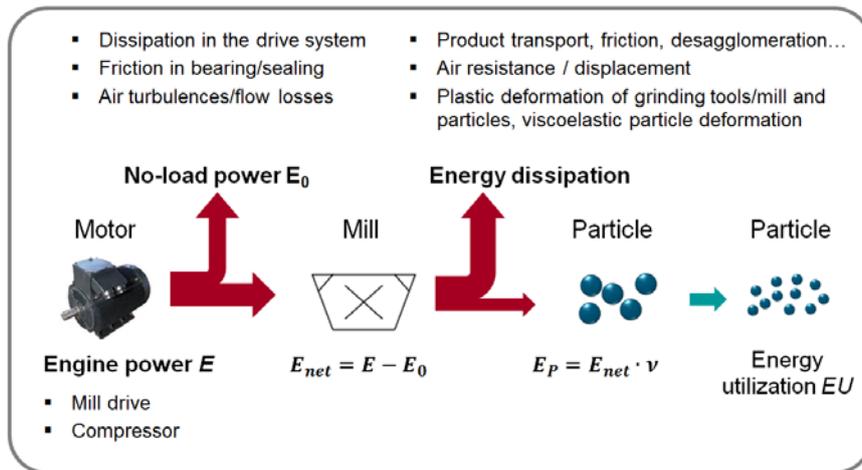
The aim of this project is to develop a system engineering approach for optimizing and scaling industrial dry fine grinding processes, with a special focus on the manipulation of the material by grinding aids. Thus, the approach will not only cover common parameters such as the mill type, process parameters, plant design or material as well as energy flows. In particular, the most important influences of applied grinding aids on the sub-processes inside the grinding plant are also taken into account. The developed approach can then be used as the basis for later flow-sheet-simulations of different complex industrial grinding processes. The investigations of a first 3-year-funding-period focus on the grinding aid impacts on the grinding behavior inside the mill. Within this funding period, the focus is put on two exemplary grinding principles, namely dry grinding in tumbling ball mills as well as impact milling in high-speed pin mills. The stressing conditions provided by these different mill types are investigated by means of DEM simulation. The breakage behavior of single particles will be determined experimentally for two different inorganic model materials, which are selected according to the demand of the IFRPI members. In addition to the breakage behavior, the particle and powder behavior between (or near) the grinding tools will be characterized by model experiments in order to describe the impact of the product formulation on the particle stressing. Together with functional grinding models and population balance models, these information will be used to predict particle size distributions and energy consumptions. For validation, grinding experiments will be performed in mills of different types and scales. In a last step, the energy utilization will be characterized as a function of both the milling conditions (mill type/geometry/size as well as grinding parameters) and the product formulation. The aim is to identify possible criteria to transfer grinding aid application to other mill types or optimize grinding aid application while scaling up processes. In a second 3-year-funding-period, the system engineering approach will be extended to continuously operated grinding processes.

## **2. First 3-year-period**

The studies within the first funding period focus on defining a general system engineering approach as well as the investigation of grinding aid impacts on the particle stressing and the grinding performance inside the mill. Following work packages are planned:

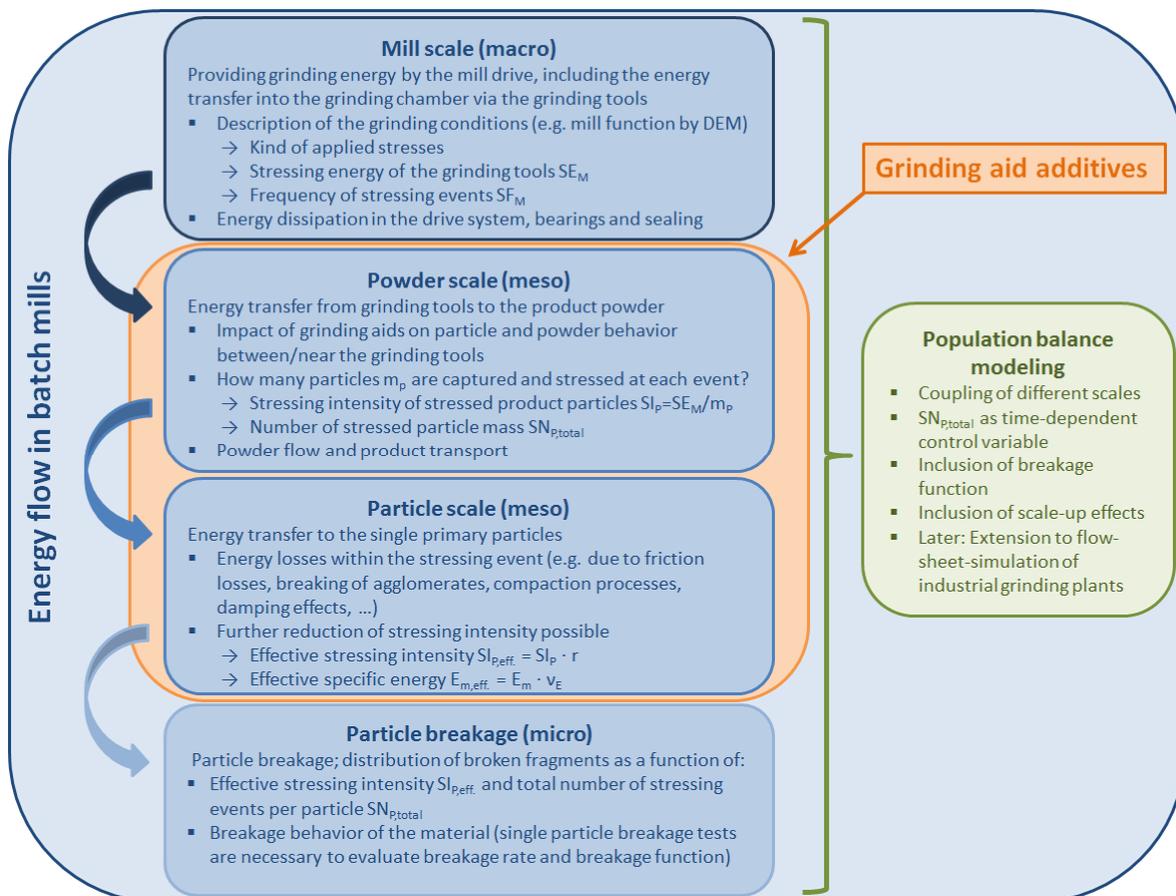
### **2.1. Development of a System Engineering Environment for dry fine grinding processes**

In a first instance, a system engineering environment is developed for characterizing simple lab-scale dry grinding processes. The approach is based on a model representation regarding the stresses acting on a particle as well as the energy flow inside a grinding machine. Similar to the approach of Kwade for wet grinding processes [1], the energy transfer from the mill drive to the particles as well as the associated energy dissipations inside the milling chamber (described by the energy transfer factor  $v_E$ ) will be evaluated according to the scheme in Figure 1. The utilization of the energy transferred to the particles is determined by the applied stress intensity, which should have a value that the particles just break. Also, the systems engineering approach will be used for modeling grinding processes with a special regard on the two different mill types. Therefore, data and findings from the different scales need to be coupled and included into a population balance model. Appropriate grinding models for the two selected grinding principles are selected based on own studies and studies from the literature. For each grinding principle, one mill related model (describing stressing energies and frequencies provided by the mill) and one breakage model will be selected in order to describe grinding conditions inside the mill as well as breakage behavior of the material, respectively.



**Figure 1:** Scheme of energy flow in mills.

For meeting the special requirements of this project, it is especially considered how far the product formulation may influence the individual sub-processes in those grinding machines and how these effects can be embedded in the system approach and the later population balance modeling (see scheme in Figure 2). Moreover, grinding-aid-specific scale-up effects will be identified and – if necessary – included in the approach.



**Figure 2:** Considerations of grinding aid effects for the system engineering approach.

## **2.2. Macro scale: Determination of the stressing conditions in different mill types**

Within this working package, the stressing conditions provided by the two different mill types are evaluated based on Discrete Element Method (DEM) simulations. Parameters like stressing energy distribution and stressing frequency are essential to feed the mill related model of the system engineering approach. Within this project, two different concepts are chosen to describe the stressing conditions for the two different grinding machines:

DEM-Simulations of the grinding ball motion are carried out for the two tumbling ball mills of different scale. The milling process will be simulated for selected parameters (e.g. mill speed, grinding ball size/material/filling ratio) that are chosen based on appropriate settings from grinding experiments from working package 2.5. The parameters will further be held constant for both mills. The outcome of the DEM study is mainly the frequency distribution of stressing energies for different kinds of stresses (normal stress, shear stress ...). Since only the grinding balls are simulated due to computing capacities, appropriate DEM input parameters need to be chosen to describe possible influences of the product particles on the restitution and friction behavior of the mill load. In a first instance, the DEM input parameters for the simulation (e.g. coefficient of restitution or friction coefficients) are chosen based on results obtained from previous studies. However, the results of the material motion which was also determined in working package 2.5 will also be considered. Thereby, it will be identified whether grinding aids may change the motion behavior of the mill load to such an extent, that the stressing behavior provided by the grinding balls is influenced by the product formulation significantly.

The stressing conditions inside the high-speed pin mills UPZ40 and UPZ100 are also estimated based on DEM simulations. However, instead of own DEM simulations, data from previous IFPRI projects as well as studies from the literature shall be applied within this project. For instance, impact velocity distributions as well as collision frequencies for grinding with the pin mill UPZ100 at different rotation speeds are estimated based on current IFPRI project "Milling and Material Grindability" [2]. For instance Ooi et al. determined the number of stressing events as a function of the impact velocity within the UPZ100 by means of DEM for different feed rates and rotary speeds. A down-scaling of these parameters to approximate the stressing conditions inside the smaller UPZ40 is carried out using the scaling-approach of Juhnke [3]. Since the motion of the rotor and the stator inside pin mills is independent of the product formulation, the impact of grinding aids on the mill function is negligible at this point.

## **2.3. Meso scale: Systematic investigation of grinding aid impacts on the particle stressing**

According to the above mentioned findings from own investigations and studies from the literature, the interactions between grinding aids (including their influences on the product properties) and the stressing conditions provided by the mill have to be well-known for an effective selection of appropriate grinding aids (e.g. [4-9]). Therefore, different basic laboratory tests will be performed to investigate the particle stressing for different mill types. The experimental setup comprises the grinding aid influences on the powder properties as well as the particle and powder behavior inside the mill, but especially between or near the grinding tools.

Even though the actual breakage behavior of single particles is not affected by grinding aids (e.g. [10-13]), the stressing behavior of the material is strongly influenced by the presence of these additives. The reason for that is the change of the particle and powder behavior as a consequence of adsorbed grinding aid molecules. It particularly changes the behavior of the material between or near the grinding tools, leading

to a change of the stressing intensities and stressing numbers. Consequently, two important aspects have to be well known to estimate the impacts of grinding aids on the stressing behavior: First, the impacts of the additive on the powder properties need to be characterized. Secondly, rules, in how far these properties determine the material behavior between or near the grinding tool under realistic and dynamic conditions, should be derived. Thereby, also the energy transfer coefficient  $v_E$  which describes the proportion of the energy being finally transferred to the product particles needs to be estimated and considered. To investigate these phenomena, following tests are included:

On the one hand, the impacts of three different typical grinding aids (alcohol amine, glycol, carboxylic acid) on selected particle and powder properties of the model materials are determined. Therefore, both model materials will be evaluated in three different, comparable size fractions. Prior to the characterization, the grinding aid molecules will be dry-mixed to the different size fraction using a plug-shear-mixer. For each of the three grinding aids, one identical and appropriate grinding aid concentration in the range of typical industrial applications is chosen. After dry mixing, the grinding aid effects are analyzed by measuring the powder flowability with a Schulze ring shear tester, the tendency of agglomeration by dry particle size analysis as well as the fluidization behavior by powder rheometer measurements. On the other hand, the same powder samples will be used for evaluating the particle stressing between the grinding tools.

The experimental characterization of the particle stressing under real stressing conditions is highly time-consuming and exceeds the scope of this project. Since similar investigations are the subject of a parallel iPAT-project, only exemplary tests using the selected model materials will be carried out within the IFPRI-project. For evaluating the stressing behavior in media mills, two different tests will be used: First, a recently designed experimental set-up based on a drop-weight-tester is chosen to investigate the particle behavior between rapidly approaching grinding balls. Thereby, especially the particle capturing between the balls will be investigated as a function of the feed size and the applied grinding aid. Secondly, a modified Hardgrove test will be used to characterize the particle behavior below rolling grinding balls, which is also an important mechanism that contributes to the particle stressing in tumbling ball mills. Both tests will be evaluated by mass balances and high speed videos. The results are used to estimate the mass of material that is stressed at each stressing event. This parameter is highly important to derive a relation for describing the actual stressing intensities  $SI_p$  (how much energy is transferred per mass of particles at each stressing event) and product related stressing numbers  $SN_p$  (how often is each particle stressed within the process) as a function of both, the stressing conditions provided by the mill as well as the product formulation. Also, the formation of strong agglomerates as well as of material coatings on the balls are evaluated in order to identify critical processing conditions, where the particle stressing between grinding balls could become less efficient due to both, energy losses while breaking agglomerate structures as well as damping effects caused by adhesion layers on the balls. Aspects like this will be considered for describing further energy losses, and thus, calculating the effective stressing intensity  $SI_{p,eff}$ . In contrast to media mills, the stressing conditions inside pin mills are assumed to be less dependent on the powder behavior. Here, the stressing intensities  $SI_p$  and product related stressing numbers  $SN_p$  are rather determined by the process parameters such as pin geometry and rotary speed. However, grinding aid effects are still possible in pin mills, since the formation of strong agglomerates and product adhesion on the pins may reduce the effective stressing intensity  $SI_{p,eff}$ . Therefore, the surface energies of the model material will be measured by means of inverse gas chromatography as it was done in a previous study [14]. The surface energy values will then be used to calculate the strength of agglomerate structures towards

impact stresses (compare e.g. [15]) and correlated with the size reduction efficiency of the actual primary particles, which is determined from the grinding experiments within working package 2.5. Moreover, air-jet-supported dry dispersion experiments (similar to the Scirocco procedure introduced by Bonakdar et al. [16]) will be performed to investigate how far the extent of agglomeration is not only determined by the product formulation but also by the dry dispersing intensity. Thereby, the extent of agglomeration is analyzed by measuring the dry particle sizes in air flows of different velocities by laser diffraction. The aim is to identify whether the grinding aids help to disperse the particles in a dry and turbulent environment, which may lead to an increased impact stressing of single particles instead stressing of agglomerates. Using this method as a real breakage tester – as it was originally used by Bonakdar et al. for soft organic products – is hardly possible in the present case: Most likely, the stressing intensities inside the dispersion unit are not high enough to enable breakage of the primary particles of these fine and comparatively hard inorganic materials.

#### **2.4. Micro scale: Investigation of the breakage behavior**

The breakage behavior of single particles is measured by a two-roller-tester. Since the breakage behavior of single particles is independent of grinding aids for realistic grinding conditions (e.g. [10-13]), these tests are only performed for the raw particles without any additive. For both model materials, particles of different sizes between 30 – 300 µm are analyzed in order to determine breakage rates and breakage function under defined stress loads. These breakage data will then be used as input parameters for the breakage model, which is embedded in the system engineering approach. Furthermore, experimental tests using a modified “picozirk” impact unit (Hosokawa Alpine) will be used to validate the influences of the grinding aids on the impact stressing of the model materials.

#### **2.5. Grinding experiments**

Within this working package, a number of dry fine grinding experiments will be carried out for the two selected model materials. Thereby, both milling principals (tumbling ball milling as well as impact milling) will be investigated using two mills of different scale each. Within these tests, the specific energy consumption as well as the product particle size distribution will be evaluated.

For ball milling, two different batch-wise operated tumbling ball mills with volumes of 4.4 L batch mill and 60 L batch mill will be used. Different grinding times will be applied to realize grinding in a wide range of specific energies as well as product finenesses. The milling will be carried out under selected process parameters for both mills. The focus of this investigation is also put on the impacts of the different grinding aids on the macro processes inside the mill. Besides the grinding result and the energy consumption of the mill, the motion behavior of the mill load will be evaluated while milling with different grinding aids. Thereby, the side plates of the drum will be equipped with transparent lids to enable the investigation of the material motion by high speed cameras.

Impact milling will be performed in a high-speed pin mills, either UPZ40 or UPZ100 (Hosokawa Alpine). Thereby, the stressing intensity will be varied by different circumferential speeds of the wheel. For the experiments, the energy consumption of the motor unit as well as the product particle size distribution will be measured. In order to obtain also very fine material in the lower micron range (thus the size range, where particle-particle interactions become more decisive), the product material will be processed in several grinding cycles. Since impact mills have comparably low mixing capabilities, the grinding aids are

dry-mixed to the feed material prior to the grinding, instead of being added directly into the grinding chamber.

For all grinding experiments, the amount of product adhesions on the mill equipment will be determined for different specific energies. While for the ball milling mainly the adhesion on the grinding balls will be taken into account, the material coatings on the pins (rotor and stator) are evaluated for the pin mill. Thereby, adhesion-induced poor stressing conditions caused by damping effects at the coated grinding tools can be identified and correlated to parameters like product fineness and product formulation. If necessary, a these effects will be considered within the effective stressing energy (see above).

## **2.6. Population balance modeling**

Since the size distribution of the final ground product is of major interest for this project, Population Balance Models (PBM) instead of simplified grinding models need to be incorporated in the present system engineering approach. Therefore, already existing PBM approaches will be used and coupled with the characteristic stressing parameter, especially the frequency distribution of stress energy, obtained from the DEM simulations. In the present case, these so-called PBM-DEM approaches are further extended with the findings from the model experiments on the particle stressing, in order to depict the impacts of the particle and powder behavior, and thus, a more realistic grinding environment. Furthermore, the energy transfer factor will be embedded to include further aspects of energy dissipations within the single stressing events.

For instance, a procedure similar to the mechanistic ball mill model of Tavares and co-workers [17] will be used as the basis for the ball mill modelling. As mentioned above, breakage rates and function for single particles can be taken from the two-roller-test. Stress loads can be estimated by the stressing intensities  $SI$ , which is the critical variable within this model. This parameter will be calculated by the stressing energies inside the mill obtained by the DEM simulation as well as the active stressed particle mass  $m_A$  per stressing event, which is approximated by a model developed from the model experiments. The product related stress number  $SN$  will be embedded as the time-dependent control variable with the population balance model. The populations balance modeling will allow a relation of product particle size distribution and energy consumption as a function of both, milling conditions as well as product formulation.

## **2.7. Development of transfer and scale-up criteria**

Within this working package, the grinding aid effects will be evaluated with a special regard on differences between mill type and mill size. All considerations are based on the systemic investigations from the shown studies. The aim is to identify, how far grinding aid applications depend on the applied grinding principal and whether it is possible to transfer them to other grinding processes. Also, aspects of adjusting grinding aid application while scaling up grinding processes are taken into account.

For enabling such a procedure, the actual energy utilization is considered as a function of both, machine related parameters and product formulation. Therefore, the total amount of specific energy, which is introduced into the grinding chamber over the processing time, is related to the actual amount of energy, which is finally transferred to the product particles and can be thus used for breakage. It is then estimated which aspects of energy dissipation depend on both, grinding aid application as well as (geometrical) scale effects. Finally, the above-mentioned energy transfer factor, which considers all grinding aid as well as scale dependent energy dissipations, will be calculated for each of the two grinding processes that were

investigated. This parameter will then be examined for its suitability to be used as critical values for scaling and transferring grinding aid applications. Thereby, it is also evaluated how far the configuration of the particles during stressing (single particle vs. multi-particle stressing vs. particle beds) determines the energy transfer.

### 3. Second 3-year-period

As already mentioned above, the system engineering approach for the ball mill will be extended to continuously operated grinding processes in a second 3-year-funding-period. Based on this approach, a flow-sheet environment for modeling industrial grinding processes will be created. The aim of the flow sheet simulation is to model whole continuously operated grinding plants (in open- as well as closed-circuit mode) for different material/grinding aid combinations. The investigations within this funding period will first of all focus on continuous grinding processes of inorganic materials in ball mills, and will be based on the materials, processes and results of the first funding period. Also, a transfer of this approach to grinding of organic materials will be evaluated experimentally. Therefore, we recommend the use of a pilot scale impact mill, with or without an additional air classifier. Upon request of IFPRI, the developed approach can also be validated for other mill types such as dry operated stirred media mills. This of course would reduce the scope of the other investigations.

In a first instance, the impact of different grinding aids on the material transport through a ball mill in open circuit mode is investigated. Thereby, the residence time distribution will be determined experimentally in order to enable the calculation of the total number of stressing events per product particle within one grinding passage. These essential information will then be included in the system approach by transport models, which are extended with parameter describing the powder flow attributes to enable an inclusion of the grinding aid effects. Then, the system approach is extended with further models for describing the air classifier performance. As a basis, the impacts of grinding aids on dynamic air classification are also identified experimentally. The corresponding effects are included in the approach by classification models, which also consider appropriate powder characteristics (such as the agglomerate strength or the fluidizability) to include grinding aid effects.

This extension of the approach is then validated by experimental grinding aid studies in an exemplary closed-circuit grinding process. A detailed outline of the proposal for the second funding period will be submitted later.

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