Mill Selection and Optimization of Size Reduction of Soft Materials via a Knowledge-based Approach based on the Determination of the Material and the Machine Function via Single Particle Stressing Experiments

Research proposal submitted to the International Fine Particle Research Institute (IFPRI) submitted by

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Introduction and problem statement

For realization of breakage, the elastically stored energy within the material has to be higher than the critical energy for crack propagation [1]. The elastic energy provided for breakage is introduced into a particle typically via its deformation by external forces in a stressing geometry defined by the type of the employed mill. The outcome of a stressing event, i.e. if a particle will break or not, depends on the interactions between the mill and the particle, which can described by a *mill function* and a *material function* [2]. Knowledge of the mill function and the material function for various materials and types of mills will allow for a knowledge-based choice of the optimum milling technology for the respective materials as well as the prediction of optimal process parameters in view of desired product properties (e.g. size (and shape) distribution, degree of disintegration of complex particles, mechanochemical activation) and process efficiency (cf. necessary mass-specific comminution energy, process time). While it has been demonstrated that for the comminution of brittle materials, like glass beads, oxides, but also organic crystals or amorphous (brittle) thermoplastics, the milling process can be successfully deconvoluted in a mill function and a material function [2–4], it is a somehow open question to which extend this approach can be transferred to 'soft materials', which often show visco-elastic, plastic or ductile material behavior. The lack of a comprehensive understanding, how soft (ductile) materials relevant for many industries deform and break, and what are the consequences for mill selection and choice of operating conditions, was identified at the IFPRI milling workshop held in Carry-le-Rouet this June. This resulted in a project brief "Mill Selection and Optimization for Size Reduction of Soft Materials", which is addressed by this proposal. Soft materials to be included as possible materials for the proposed study include food powders (e.g. from grains or legumes (starch and/or protein flours), sugar, cocoa (chocolate), coffee, milk powder, spices etc.), biopolymers, polymers (e.g. engineering thermoplastics), organic composites and 'complex particles' (e.g. multi-component granules / inorganic loaded agglomerates).

In contrast to brittle (elastic) matter, for example, viscoelastic materials are characterized by a complex Young's modulus, i.e. depending on the stressing conditions a significant portions of the total introduced energy is not stored elastically, but subjected to viscous dissipation and, consequently, not available to initiate crack formation or to promote crack propagation. Moreover, their mechanical responses exhibit rate dependencies, i.e. the breakage will depend on stressing velocity. Moreover, non-brittle materials like semi-crystalline engineering thermoplastics, that have been studied in wet grinding in stirred media mills [5–7] in own previous work, do not only show way slower, but also 'unusual' breakage kinetics (with an induction period until fracture sets in, see Figure 1, left) as compared to what is known on breakage of brittle materials like oxides, sulfides or ores in this type of mill.

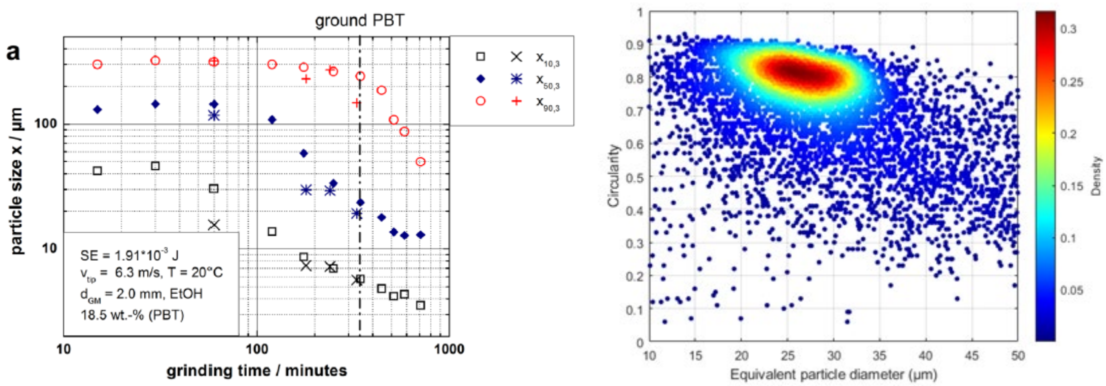

Figure 1: Evolution of product particle size with process time for wet comminution of polybutylene terephthalate – breakage kinetics are slow and 'unusual' [7] (left); 2D distribution (circularity vs size) of impact comminuted pea fragments: product quality in view of starch globule and protein globule liberation can be assessed by these representations (unpublished own work) (right).

The *machine function (mill function)* depends on the used type (and geometry) of the mill, which determines the type of stresses (cf. compression, shear, impact, cutting stress) acting onto the particles, as well as the chosen process parameters, that determine amongst others the (distribution of) stress energies and respective stress frequencies within the stressing zone of the mill. For example, roller mills will provide (two-sided) compressive stress (at moderate stressing velocities and shear stress), while jet mills or pin mills will provide (one-sided) impact stress at high stressing speeds (but rather small stressing frequencies). Media mills, like e.g. the typically wet-operated stirred media mills, will provide compressive and shear stress and are characterized by a rather high stress frequency and higher stress numbers [8] as compared to the aforementioned dry grinding approaches. In wet grinding in stirred media mills, in addition, the employed dispersing medium will remarkably influence the stressing conditions. For example, the stressing zones, the flow within the grinding chamber and the collision characteristics depend on the rheological characteristics of the fluid [9, 10]. The transferable stress energy available for fracture in a media mill can be significantly reduced due to viscous dissipation [5] and the active volume between the grinding beads, where stressing of particles may take place, is influenced by rheological characteristics of the fluid phase via the viscosity-dependent displacement flow and subsequent particle displacement between approaching grinding media (reducing the probability that the particle is stressed) as well.

It becomes obvious, that the determination of the *mill function* can become complex, but feasible 'tools' to tackle this task, i.e. simulations (e.g. DEM, Monte Carlo, CFD, coupled DEM-CFD), but also experimental approaches based on *probe particles being e.g. responsive on the magnitude of the applied energy* and/or number of stressing events [11, 12] (own work) exist and have been proven as useful.

The *material function* describes the material's response to stress and strain and under which constraints material failure will occur. Thus, a proxy for the material function can be the change in PSD when subjecting a feed material to certain stresses. But also the obtained product shape distribution (see Figure 1, right) will depend on the type and magnitude of the applied stress. The material function depends generally speaking on the material law and the mechanical properties, like e.g. the (complex) Young's modulus, Poisson ratio, fracture toughness, yield stress or elongation at break, and how stress and strain are applied (cf. magnitude, strain rate, one-sided vs. two-sided stressing, compression vs. elongation, shear). Moreover, it will depend on particle size (cf. smaller particles are harder to break as expressed in the Bond, Kick and Rittinger laws [13]). There

will be a certain distribution of outcomes of a stressing event, if (nominal identical) particles are stressed under identical conditions with respect to breakage, i.e. a breakage probability exists. Amongst others, this is due to the (random) distribution of flaws within the materials or the presence of already existing cracks.
Vogel and Peukert [3] were able to show that the breakage probability of brittle materials follows a Weibull distribution. The *breakage probability* can be determined experimentally from *single particle breakage experiments* under defined conditions, i.e. known introduced energy (and stressing history) and defined stressing geometry. The obtained experimental data can be used as input for breakage models like e.g. those by Vogel and Peukert or Ghadiri and Zhang [3, 14, 15], respectively, as basis for the adaption of existing breakage models, respectively, development of new model pictures.

Goals and addressed research questions

In view of the problems statement the main goals addressed in this project are to
(1) gain a deeper *understanding how soft materials deform and break* and, consequently,
(2) which *milling technology* should be applied under which *process parameters* to
(3) obtain a *product of desired properties* under given constraints (necessary mass-specific comminution energy consumption, process time, fines content etc.).
The 'desired product properties' are not understood as 'only' the particle size distribution, but should also include other particle and bulk solid characteristics like particle shape distribution, which may give valuable information on the mode of breakage when combined with PSD data, respectively, can be crucial for certain applications. Also changes of material properties due to stressing and breakage, such as changes in molecular weight (of polymers) or amorphization, respectively mechanical activation [16] are in scope of the study (and could be monitored with the available techniques at the institute). Here input from IFPRI members would be helpful, on which material properties in addition to particle and bulk solid properties the project should focus.
The project aims to achieve the above mentioned three goals and aims to answer the following fundamental research questions based on a (mainly experimental) approach based on a combination of single particle breakage experiments with material characterization (to determine the material function) with determination of the mill function (machine function) by load-sensitive probe particles, milling experiments (parameter studies) and the development (or adaption) of breakage models:

- To which extend need established breakage models be adapted to describe the breakage of soft materials and under which conditions is this adaption feasible?
- If the adaption of established breakage models fails: what are the (most) relevant mechanical properties that need to be included in novel breakage models for soft materials?
- In contrast to brittle breakage, for breakage of viscoelastic, plastic and ductile materials there will be a pronounced 'path dependency'. How can this effect be incorporated into breakage models (and under which constraints are simplifications feasible)?
- How feasible is an extension of (established) breakage models not only with respect to incorporation of the description of the breakage probability or fragment size distribution of soft particles, but also with respect to other characteristics like e.g. particle shape distribution or other product properties (molar mass degradation in polymers, amorphization, activation)?

Model systems

Within this project three (to four) well-defined model particle systems for soft materials, which have relevance in the industries, shall be considered. It is proposed to perform the studies for
(1) polybutylene terephthalate (PBT), a semi-crystalline, ductile thermoplastic, where besides using mm-sized (commercially available) granules also (close-to) spherical model particles (up to max.

100 to 130 µm size) are available [17] (own work, process also available for other thermoplastics). Moreover, the studies shall be extended to

(2) filled polymer systems, which e.g. are used as toner systems. The incorporation of variable amounts of (inorganic) fillers into (close-to) spherical particles is possible with the approach proposed for the PBT in (1). This model system shall address inorganic loaded systems or composites mentioned in the brief. Finally, as model systems for food powders

(3) legumes (peas (or lentils) < 4 mm) shall be used. These are narrowly distributed 'multicomponent, complex particles' of defined shape and they are of interest in optimization of the liberation of their protein bodies in view of applications for meat substitutes. Additionally,

(4, option to be discussed) milk powder could be considered to be included as a model system for 'food agglomerates / complex granules'.

It is important, that the model particle systems are available in a well-defined shape (ideally spheres), because shape can affect breakage probability. Also here, the input of IFRRI members and their opinion on the proposed systems is appreciated. The model systems mentioned are not yet finalized, but are proposed and may be adapted following discussion before the project begins. Moreover, industrially relevant samples of IFPRI members shall be included in the studies (also here, the systems to be investigated have to be discussed).

Methodology

Single particle breakage experiments

To achieve the above mentioned goals, the *material function shall be determined via experimental approaches* based on *single particle breakage experiments* under defined conditions using a single particle impact device that allows for (one-sided) central impact proposed by Schönert and described e.g. in [4] (see Figure 2, left).

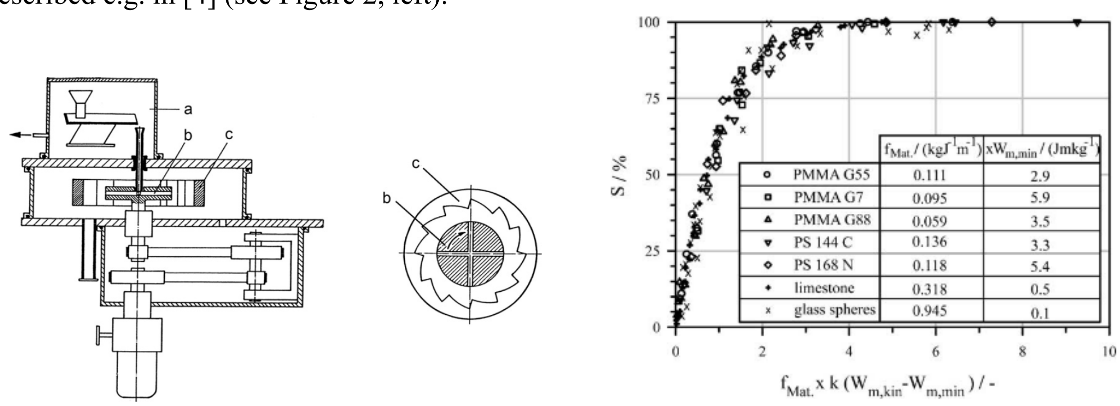

Figure 2: Single particle impact device proposed by Schönert (taken from [4]) (left), breakage probability S of different materials as described by a master curve according to the breakage model by Vogel and Peukert [3] (right). See text below for discussion.

In this device, the particles to be tested are centrally fed into a rotor that is equipped with channels. The particles are accelerated in these channels and exit the rotor at a defined (known) speed that can be calculated from the rotor speed (and also be expressed as a mass specific kinetic energy $W_{m.kin}$). Due to the special shape of the outer ring, where they impact, it is guaranteed that central impact applies for any trajectory the particle can follow after exiting the rotor channel. The device can be evacuated, which allows the reduction of drag forces acting on the particle allowing also for stressing of small particles (in the range of several 10 microns). These *one-sided (central) impact breakage experiments* can be supplemented with an impaction device (particles are accelerated by a jet expanding in an evacuated chamber with an impaction plate), that allows for *oblique impact*. From single particle experiments, parameters for breakage models can be extracted. For example, the model by Vogel and Peukert [3] accounts for the material behavior by two parameters: f_{mat}, that describes the resistance of a material against fracture and $W_{m,min}$, a characteristic mass-specific energy that can be introduced into a material without causing fracture. The breakage probability S, see Figure 2, right, was found to follow a Weibull for many brittle materials subjected to impact:

$$S = 1 - exp\left(-f_{mat} \cdot x \cdot k \cdot \left(W_{m.kin} - W_{m,min}\right)\right)$$

Here, all quantities besides the already mentioned two parameters describing the material's response to stressing, f_{mat} and $W_{m,min}$, are known, or can be set or measured, like k, the number of impact events, or x, the particle size (known from PSD of the feed material to be stressed), $W_{m.min}$ (particles do not yet break at that energy input) and finally S, which is accessible via comparison of the PSDs of the feed and the product.

In addition to impact breakage experiments, for material systems that are known also to break under *(two-sided) compressive stress*, single particle stressing experiments under defined varied normal loads will be performed. These experiments will be possible for larger (mm-sized < 4 mm) particles and will also allow to get insights into the stress-strain behavior in this size range. These experiments are planned to be supplemented by single particle stressing in a *horizontal roller mill testing device* (allowing to add shear stress to the compressive stress [18]), that besides variation of the rotation speed also allows to vary the gap size between the two counter-rotating rollers. Breakage models can be applied in a similar way as described above.

Characterization of stressing conditions in mills by means of probe particles

An experimental approach towards the *determination of the machine function*, i.e. which stresses (distribution of stress energies and stress numbers) a mill transfers to a material is the *application of probe particles being sensitive on mechanical stimuli*. The direct measurement of stress energy and stress numbers in stirred media mills [11, 12] and jet mills [19] has been performed in previous work at the Institute of Particle Technology applying spherical copper particles of several 10 microns size as probe particles. Copper as a ductile metal deforms upon stressing irreversibly. Thus, these particles are ideal probes for monitoring the introduced energy upon stressing. It was shown that the introduced energy can be determined by FEM modelling (cf. simulation of a compression experiment) by reproduction of the change of the shape of the probe particles accessible experimentally via image analysis of optical microscopy and/or electron microscopy images [11]. In addition, the number of impact events can be determined from image analysis of the stressed particles by counting the number of flattened areas. Further details on the procedure, which shall be applied in the studies of in this proposal, can be found in the papers by Strobel et al.[11, 12, 19].

Comminution experiments / parameter studies

For the determination of the breakage of the model particle systems and the industrially relevant samples provided by IFPRI members, comminution experiments will be performed in mills in batch mode. Process parameters will be varied and samples will be drawn over the process time to monitor the response of the model particles to stressing and their breakage. The *probe particles will be used to determine the machine function under the stressing conditions the model particles have been subjected to*. It should be possible to describe (and predict by means of a suitable model) the observed outcome of the comminution experiment by combining the material function determined in single particle breakage experiments with the mill function determined by means of probe particles. The comminution experiments will be used for the development of breakage models, respectively, for their verification (or falsification and subsequent improvement).

For the experiments, impact mills such as a (lab-scale) rotor mill Pulverisette 14 (Fritsch and a jet mill (AFG100, Hosokawa) are available. In this project the rotor mill (in pin mill configuration) and the jet mill (in batch mode) shall be employed. For materials that also can be comminuted by compression, a roller mill shall be employed as well.

Proposed work program and time line

The work program given below is designed for a project duration of three years (12 quarter years, Q). To achieve the goals employing the methodology outlined in the previous section to gain a deeper understanding of breakage of soft materials by determination of the material and the mill function, performing parameter studies in mills and developing breakage models, the project is structured into five work packages (WPs):

WP 1: particle and material characterization
This is the central characterization WP of the project. Here, the model systems (see respective section), the copper probe particles for determination of the mill function by the approach described in the methodology and in [11], and the materials provided by the IFPRI members as well as the stressed particles from WPs 2 and 4 and the comminution products from WP 3 will be thoroughly characterized with respect to size and shape distribution and surface morphology (employing laser diffraction particle sizing optical and electronic microscopy) and bulk solid characteristics (flowability via shear testing, packing density). Also the mechanical characterization (Young's modulus, Poisson ratio, elongation at break, stress-strain behavior of specimen) will be done in this WP. Further materials properties that might change due to stressing and fracture like rheological, thermal or optical properties (rheometry, DSC, spectroscopy) of the materials will be characterized here as well.

WP 2: determination of the material function (single particle breakage experiments)
In this WP single particle impact and compression experiments with the model particles (and the systems provided by IFPRI members) will be performed in the test devices described in the methodology section entitled "Single particle breakage experiments". The obtained stressed particles and fragments will be characterized in WP 1. In the impact stressing devices, impact energy (i.e. impact velocity) and geometry (central vs. oblique impact) will be varied, in the roller mill test device, roller speed and gap width. Particle fractions will be stressed consecutive times to get insights into the dependence of the number of stressing events and size on breakage. Moreover, for single particles experiments that will allow to assess possible 'path dependencies' of the fracture outcome will be performed (cf. stressing first with high energy, then with lower energy vs. first stressing with lower, then with high energy). The particles are stressed at known energies and with known modes of stress and at known geometry. Thus, combined with the characterization from WP 1 the material function can be established (which will be used for breakage model development in WP 5).

WP 3: milling experiments / parameter studies
Milling experiments with the model systems (and the samples from IFPRI members) will be conducted in this WP in parallel with the single particle breakage experiments in WP 2. A parameter study under variation of typical process parameters for the respective mill will be performed. Mills used here will be a pin mill, a jet mill and a roller mill. Products are characterized in WP 1.

WP 4: determination of the mill function by probe particles
Milling experiments will be performed with the copper probe particles, respectively, with mixtures of model particles (and systems provided by IFPRI members) and the probe particles in the mills described in WP 3 under the process parameters chosen there. Evaluation of the shape change of the copper probe particles (WP 1) as described above and in [11] combined with FEM modelling (done in this WP) will allow to get insights into the acting stress energies and stress numbers and, thus, the machine function can be determined.

WP 5: adaption of existing or development of new breakage models for soft materials
In this WP all the results of the previous WPs are consolidated and breakage models are established. It is evaluated, if established breakage models like those mentioned in the methodology can be applied to breakage of soft materials, respectively, how these models have to be adapted. It will be evaluated, if the outcomes of comminution experiments from WP 3 can be rationalized with the

model based on the machine function and the material function from WPs 4 and 2, respectively, it will be evaluated how predictive the breakage model is. In addition to the predictiveness regarding which PSD is obtained under which stresses, the possibilities to extend the model predictiveness to other product features like shape distributions shall be evaluated in this WP. A possible approach could be the development of models based on principal component analysis / correlation analysis.

The time line is given in the following Gantt chart. It is planned that there is the possibility for an iteration loop. After year 1 (Q 4) the whole approach should have been established and have been shown to work for two of the model systems. Systems provided by IFPRI members shall be included in the studies from Q 5; from Q 9 only studies shall be performed with these materials.

work packages	Q1	Q2	Q3	Q4	Q5	Q6	Q7	Q8	Q9	Q10	Q11	Q12
WP 1	■	■	■	■	■	■	■	■	■	■	■	■
WP 2	■	■	■			■	■	■		■	■	■
WP 3			■	■	■			■	■	■	■	
WP 4		■	■			■	■	■			■	■
WP 5			■	■	■		■	■	■		■	■

If the project works as planned, we will get new insights into the breakage of soft materials and we will have obtained machine and material functions being of interest beyond the project that can be used by IFPRI for follow-up projects.

Potential risks and mitigation strategies

Two possible main risks have been identified: (1) "The dependencies of breakage of soft materials are too complex and cannot be described by analytical model approaches like those mentioned above (or extensions)." This risk is assessed as low to moderate. If necessary, data-driven modelling could be applied, which will allow to describe the breakage. (2) "The deconvolution of a comminution process in a material function and a machine function is not possible, respectively, it is not unambiguous." This risk is assessed also as low to moderate. If necessary, data-driven modelling could be applied as well, which will allow to describe the comminution process.

Collaboration with IFPRI members

As already mentioned in the section 'Model systems', close collaboration with IFPRI members is sought well in advance of the project start date, to discuss and, if deemed feasible, adapt the proposed model materials systems (and/or used mill types). Later on in the project phase, regular discussions on the project progress and input by IFPRI members, as well as the provision of 'real' systems from the companies to be included in the proposed studies will be appreciated. The transfer and testing of the project outcomes in a joined effort, i.e. the application of determined material functions and breakage models to larger-scale milling equipment available at the IFPRI companies would be appreciated as well. Testing the applicability of the models and approaches also at larger scale and in more complex process chains will allow to evaluate them in a production environment, judge their feasibility and provide information, if refinements or adaptions are necessary.

How this project leverages existing research in our group

The project will help to fund new and interesting research in the field of comminution of soft materials in our group, that could not be performed without funding. The project will extend the competences in our group on characterization of the breakage of soft materials of different material classes beyond polymers. The project and its outcomes will provide the basis for further projects addressing processing of soft materials in more complex process chains like e.g. grinding classifier circuits. Moreover, the outcomes also will provide valuable insights in applications, such as pneumatic conveying or storage of soft materials, where breakage is undesired. Ideally the project outcomes can be used by the IFPRI partners and for follow-up projects.

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