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Validation of a particle impact breakage model incorporating impact number effect

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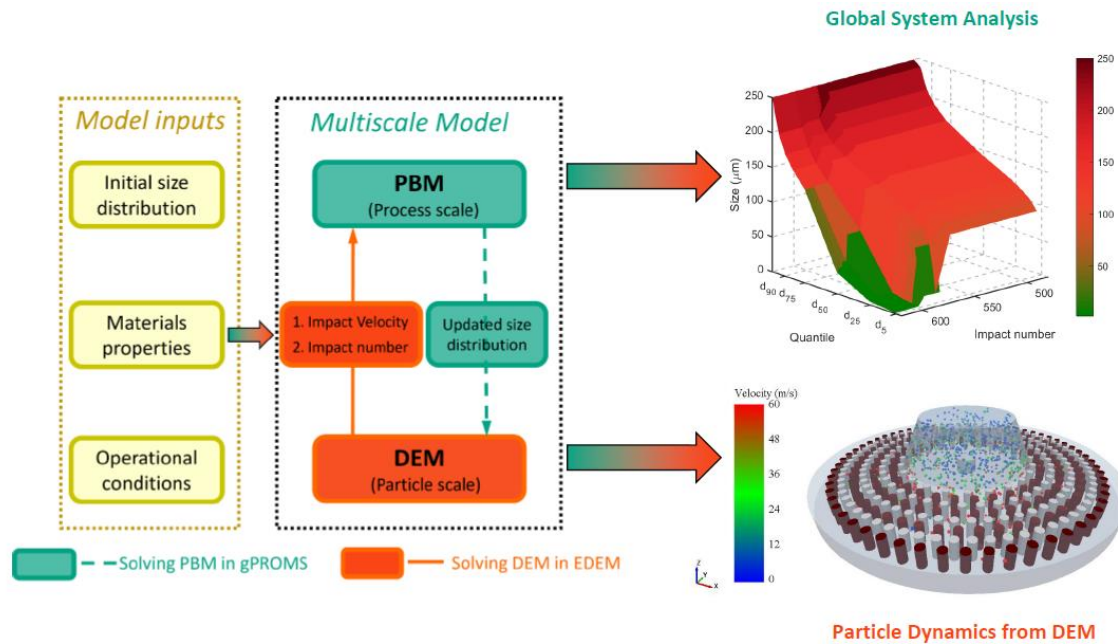
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Abstract

This paper presents validation of a particle impact breakage model i.e. Vogel and Peukert model with a focus on the impact number. The Vogel and Peukert model developed based on mechanical and statistical foundation has been widely used in various fields such as mineral engineering and chemical engineering but is barely studied in the application of repeated impact. The selective breakage data in the literature is collected to provide the database for model validation. It has shown that the Vogel and Peukert model is generally applicable to all the breakage cases considering the impact number. The effect of impact number is further elaborated in the population balance model (PBM) whereas the particle dynamics are provided from Discrete Element Method (DEM) simulation of an impact pin mill. The global system analysis of impact number is carried out with the synergic effect from impact velocity. The successful validation of Vogel and Peukert model incorporating the effect of impact number demonstrates its versatility whilst other key parameters such as impact energy and particle size can be considered in parallel.

Keywords: Model validation; Impact number; Damage accumulation; DEM-PBM coupling; Global System Analysis

Graphical abstract:



Highlights

- The effect of impact number is validated for an impact breakage model.
- The impact number in particle breakage is studied in particle and process scales.
- The impact breakage model is implemented in a DEM-PBM coupling framework.
- Limitation and extension of the breakage model are stressed for the future study.

Nomenclature

a	Fitting parameter, -
b	Fitting parameter, -
c	Fitting parameters, -
D	Degree of damage, -
d	Fitting parameters, -
d_0	Model parameters
d_θ	Model parameters
E_{50}	Median impact energy, J/kg
E_{cs}	Mass-specific impact energy, J/kg
E_{min}	Material-specific parameter, J/kg
E_n	Fracture energy on nth loading cycle, J/kg
f_{Mat}	Material-specific parameter, kg/(J m)
M	Model parameter, -
m	Particle mass, g
n	Impact number, -
P	Breakage probability, -
q	Fitting power exponent, -
r	Damage accumulation, -
t_{10}	Breakage index, -
S	Selection function, /s
v	Impact velocity, m/s

$W_{m,kin}$	Mass specific impact kinetic energy, J/kg
$W_{m,min}$	Threshold kinetic energy, J/kg
x	Particle size, mm
y	Fragment particle size, mm
y'	Minimal particle size by breakage, mm
α	Strain under the first compressive stress, -
α_c	The critical strain at the fracture point, -

1 Introduction

Particle breakage subject to stressing events is prevailing in the size reduction of a milling process [1,2]. Amongst these stressing events, impact breakage is one of the dominant failure modes in the milling processes such as ball mill [3] and impact pin mill [4, 5]. To ensure a desired size reduction ratio, a quantitative prediction of the milling performance of particulate materials is thus required. The population balance model has been widely used to predict the particle size distribution where impact breakage is overwhelming in a milling operation. The success of population balance model lies in the appropriate definition of the selection function and breakage function to describe the particle size evolution. The selection function, i.e. the breakage probability model, is used to describe the extent of particle breakage subject to impact loading. The breakage function is the mathematical function to describe the progeny particle size distribution following the impact breakage events. Notably, enormous studies of selection function have been committed in the literature to develop the fundamental understanding of the governing factors in particle impact breakage. These governing factors include but are not limited to particle velocity, particle size, particle shape, particle mechanical properties, target rigidity, particle orientation, impact angle and impact number [6–10]. To form a predictive feature of impact breakage, many impact breakage models in the context of selection function were developed where the key governing factors have been considered [11–14]. The impact velocity is generally found to be the most influential parameter in the particle breakage model. As the impact velocity is increased, inelastic deformation in the impact site results in the transition of chipping arising from lateral cracks to fragmentation arising from radial cracks [6]. The particle size is another critical factor in determining the breakage probability [15–17].

The impact breakage probability is shown to increase with the increased particle size [18]. From the perspective of energy consumption, smaller particle size requiring more specific fracture energies indicates greater energy consumption for the production of finer products in milling operation [19]. A bespoke study of the particle size on the impact breakage was carried out to correlate the particle size, material property and net cumulative impact energy with a breakage index t_{10} . The equation is presented as [20]:

$$t_{10} = M\{1 - \exp[-f_{mat} * x * n * (E_{cs} - E_{min})]\} \quad (1)$$

where t_{10} is the cumulative percentage passing 1/10 of the initial particle size; M denotes the maximum value of t_{10} subject to impact loading whereas x is the particle size; n and E_{cs} are the impact number and mass-specific impact energy; f_{mat} and E_{min} are material-specific parameters, which can be fitted from the single particle impact tests. Note that Eq. (1) is derived with the modification from Vogel and Peukert model [17] to describe the impact breakage probability, of which the details will be presented in next section. The evolution based on Vogel and Peukert model is to enable a mobilised parameter M to correlate the breakage probability with the size index in the breakage function. However, it is imperative to distinguish Eq. (1) and Vogel and Peukert model concerning the application. Eq. (1) is used to predict the broken size distribution index. In contrast, Vogel and Peukert model is widely used to predict the impact breakage probability in a wide range of impact loading events.

The validation of Eq. (1) using eight sets of drop weight impact data was performed with several kinds of ore and quarry particles spanning a wide spectrum of impact specific energies and particle sizes. Note that the Vogel and Peukert model has shown its applicability for particle size as large as 82.2 mm [20]. The validation results were shown to be effective in establishing a unified predictive curve which in large agrees well with the sourced experimental data. The successful validation is accounted by a fundamentally favourable structure of Vogel and Peukert model in capturing the governing factors for particle breakage [20]. However, the effect of impact number n in Eq. (1) and Vogel and Peukert model is usually ignored due to the fact that the majority of impact breakage tests are carried out by single impact number. Then a critical question emerging from the successful model validation of particle size is whether the Vogel and Peukert model can also be validated with regards to the effect of impact number.

This paper presents a specific study on the effect of impact number on breakage probability by validating the Vogel and Peukert model in a variety of repeated impact loading tests. The breakage database in the published literature is then collected to validate Vogel and Peukert model. The performance of Vogel and Peukert model is inspected from both the viewpoints of statistical assessment and graphical comparison between the fitting results and experimental database. The significance of the impact number in relation to the population balance modelling of particle breakage in milling operation is discussed.

2 Theoretical background

A large number of particle impact breakage models can be found in the literature and the majority of these breakage models falls in the empirical-based category [4]. In particular, the developed breakage models are mainly as a function of impact velocity or specific kinetic energy. In terms of the study of impact number and its influence on the breakage probability, single impact number is considered by default in most breakage models. With that being the case, the repeated impact loading is usually overlooked and even for the models considering the effect of impact number their applicability into a wide range of impact conditions is unclear. In the literature, there are differing notations for repetitive impact loading such as impact number, repeated loading, impact fatigue. For the simplicity and to avoid any bewilderment, all these notations are referred

with equivalent meaning and impact number is preferred in the present study. The general description of the breakage strength decay due to repeated impact loading is first presented, prior to validation of Vogel and Peukert breakage model pertinent to impact number.

2.1 Breakage strength decay

The damage accumulation resulted from repeated impact number has been presented by several researchers [21–23]. A distinct feature of repeated impact number is the weakening effect of the particle breakage strength. Assuming a particle stressed by a flat surface, the degree of damage D can be related with strain by a power relationship and it gives:

$$D = \left(\frac{\alpha}{\alpha_c}\right)^r \quad (2)$$

where α is the strain under the first compressive stress whilst α_c denotes the critical strain at the point of fracture. r is termed as the damage accumulation coefficient. The first and critical compressive strain can be measured from the load-deformation experiment. The damage accumulation coefficient r needs to be fitted from the repeated loading experiment. A lower value of r indicates a higher amenability to accumulate damage and weaken from repeated loading [22].

Analogue to the weakening effect of repeated compression, the schematic illustration of weakening effect of impact number on the particle breakage probability is shown in Fig. 1. The mechanism of weakening effect for particle strength has been explained previously [21] and it can be concluded with two postulations why a particle doesn't break for the first impact and then breaks after several stressing iterations. The first postulation is that the strength of particle depends on the impact orientation and the impact orientation favourable for pre-existing flaws initiation can be found not in the first impact but after several impact numbers. The second postulation is that the increased impact number will facilitate the growth and propagation of these internal flaws.

Fig. 1

As clearly shown in Fig. 1, the breakage probability increases with the increased impact number given the identical impact energy. This is mainly due to the progressive crack propagation of inherent cracks inside the particle, leading to diminishing particle strength with increased impact number. Notably, it has been reported that the higher energy input results in a more speedy arrival in the asymptotical maximum value of 100% in terms of breakage probability [23].

2.2 Breakage model considering impact number

Vogel and Peukert developed a breakage model on the basis of dimensional analysis and fracture mechanics and it gives [17]:

$$P_x = 1 - \exp\{-f_{Mat}xn(W_{m,kin} - W_{m,min})\} \quad (3)$$

where P_x is breakage probability; $W_{m,kin} = 1/2v^2$ (J/kg) is mass-specific kinetic energy among which v is the impact velocity; x and n are particle size and impact number. f_{Mat} and $W_{m,min}$ the material dependent properties.

In making a decision to choose a breakage model, there exists a competing mechanism between model simplicity and model fidelity. The model simplicity requires the model to be simple in the mathematical form, which is easy to interpret the functionality with minimum fitting parameters. The model fidelity requires the model to be physically sound, which is able to describe the underlying mechanisms by incorporating the critical process parameters (CPPs) such as the impact velocity and Critical Quality Attributes (CQAs) such as the fracture toughness. In this work, the Vogel and Peukert model is focused for the model validation of impact number. From the simplicity perspective, Vogel and Peukert model has only two fitting parameters and the effect of the impact number can be directly reflected in the mathematical form. From the fidelity perspective, Vogel and Peukert model was developed as a function of fracture mechanical model and statistical Weibull distribution [17,24,25]. Furthermore, as fracture roughness and hardness have been found to be key contributor in the particle breakage [15,16], the mechanistic linkage of these two mechanical properties has been linked with the fitting parameters in Vogel and Peukert model. The readers are referred for more details about the mechanistic linkage with the two fitting parameters f_{Mat} and $W_{m,min}$ [13]. However, the selection of Vogel and Peukert model does not mean a perfection in predicting the breakage probability in all possible circumstances. The practical limitation of Vogel and Peukert model should be also noted. The threshold energy $W_{m,min}$ varies inversely with the particle size and this model may not be applicable to coarser particles as their breakage probability becomes invariably constant. A comprehensive assessment of literature breakage models incorporating impact number forms a strong research interest in the future work but does not fall in the scope of current investigation.

3 Breakage database with impact number

3.1 Database collection

There are relatively few publications to specifically study the effect of impact number on particle breakage, contrasted to the effect of impact velocity and impact angles [4,26]. As the main thrust of this paper is to examine the serviceability of Vogel and Peukert model in various impact environments, it is critically important to collect the breakage database regarding the impact number as widespread as possible. However, it should be noted that the database collection is not intended to be comprehensive. Instead, only

the representative database will be collected which are used for the model validation of Vogel and Peukert model in Eq. (3).

To that end, the criterion for database collection is first proposed to ensure an unbiased approach of data acquisition. First and foremost, the breakage data of impact number in the literature excluding Vogel and Peukert model will be surveyed. Secondly, the dataset of different test materials will be compared and then the breakage probability with most experimental data points will be collected. Thirdly, the dataset including both impact velocity (or kinetic energy) and impact number will be prioritized. The dataset only with impact number will be excluded whilst the information on impact velocity or kinetic energy is unavailable. Following this criterion, the database of breakage probability with the focus of impact number is summarized in Table 1.

Table 1

3.2 Database summary

The database in Table 1 consists of 11 datasets from four publications and totals 85 data points of breakage probability ranging from 0.16 to 1.0. The impact number is varied from 1 to 19 for the tested particles including 1.4–2.0 mm NaCl, 2.0–2.35 mm Dead Sea Salt, 4.0–4.75 mm Limestone and 13.5 mm rock samples. The impact velocity is varied from 10 m/s to 27.3 m/s either by drop ball impact or single particle impact. Interestingly, the impact velocity under repeated test is not as high as that in the conventional impact tests, which is usually over 30 m/s in the fragmentation tests [27,28]. This is largely because that higher impact velocity will weaken the effect of impact number, which will make the effect of impact number less prominent compared to lower impact velocity. For the dataset based on kinetic energy, the impact velocity is equivalently converted to keep the consistency in the third column of Table 1. The breakage database is collected under normal impact and the effect of oblique impact is excluded to isolate the effect of impact number.

4 Model assessment under repeated impact

4.1 Statistical assessment

The principle of model assessment is to pick up one group of dataset for model validation whilst taking the remaining for model calibration. This is exceptional to Dataset 01, 02 and 03 with one or two datasets, which can only be used for model calibration without external validation. The fitting parameters f_{mat} and $W_{m,min}$ through Eq. (3) are summarized in Table 2 according to the breakage dataset in Table 3. It clearly shows a strong applicability of Vogel and Peukert model, as evident from a very high R^2 value. It should be noted that the optimal parameters are estimated in MATLAB using nonlinear fitting methods. For the parameter fitting with multiple datasets, the optimal values of f_{mat} and $W_{m,min}$ are estimated using a global fitting method where all the datasets including all the operational conditions are fed for parameter estimation.

The global fitting method can ensure the fitted parameters can unbiasedly satisfy the overall agreement between the predicted and measured breakage probability for all the tested datasets. This method has been successfully applied in the parameter estimation in a breakage kernel for twin screw granulation [8].

Table 2

4.2 Graphical comparison

Substituting the optimal parameters in Vogel and Peukert model, Figs. 2, 3, 4, 5 and 6 present the graphical comparison of breakage probability between modelling fitting and experimental values in the datasets. Taking Fig. 4 for example, the breakage probability predicted by Vogel and Peukert model gives rise to excellent agreement with the dataset 04, 05 and 06. Notably, the close agreements between the predicted and measured value of breakage probability can be observed in all the tested impact velocity 10 m/s, 16 m/s and 20 m/s. The similar trend of predictive closeness can also be found in the varying impact energy from the datasets 07, 08 and 09.

Fig. 2, 3, 4, 5, 6

5 Impact number in an impact pin mill

5.1 Predicting impact number in an impact pin mill by DEM

The impact number from single particle impact tests is observed with the maximum value of 19 shown in Table 1. However, it remains unclear how many impacts a particle will undergo under a realistic comminution operation. Therefore, DEM (Discrete Element Method) simulation of a full-scale Hosokawa UPZ100 pin mill is carried out to investigate the impact statistics. The reason to choose impact pin mill is that the high impact number in the mineral comminution system is less studied but bears strong scientific merit in exploring the particle dynamics and validating the Vogel and Peukert model in high impact frequency. The DEM simulations are carried out by the commercial DEM software Altair EDEM. The geometry of the impact pin mill is shown in Fig. 7 whilst more details of the impact pin mill can be found elsewhere [4,30]. The pin mill has four rings of pins fixed to a static plate and four rings of pins in a rotary plate.

Fig. 7

The raw particles are fed to the inlet from the top and the fed particles are gravitationally transported to the milling regime mounted with interlaced rotary and stationary discs. The particles will undergo considerable impact due to the high-speed rotating disk and the product will be collected from the outlet with significant size reduction. The specification of the pin layout in the rotary and stationary discs is attached in Appendix. However, it is important to note that, the distance between neighbouring pins is diminishing outwards despite the increasing number of pins in outside rings. Furthermore, the pin distance including the

pin space and net space in rotary discs are bigger than those in the stationary discs. This is due to the fact that the fed particles will be impacted by the first layer of rotary disc and then the first stationary disc sequentially.

Hertz-Mindlin contact model was used to resolve the particle–particle and particle–pin contact interactions [30]. The Hertz-Mindlin model is a non-linear elastic model and is well suited to simulate the non-cohesive interactions within the computational models. The model uses a spring-dashpot response to normal contact between particles and/or geometry and a Coulomb friction coefficient for shear interactions and a second spring-dashpot response to tangential or rolling friction interaction. This interaction model has been successfully used in the impact pin mill [30] and DEM-based particle impact breakage [31]. The numerical time is set to be 13% of the Rayleigh time step which has been previously demonstrated enough to ensure the convergence of the simulations [30]. Zeolite particles were simulated with the particle properties provided by CWK, Germany. The contact parameters between particle and pin are calibrated through the angle of repose experimental measurements. The number of inlet particles is calculated based on a feed rate of 24 kg/h and four operational conditions were simulated subject to rotary speed at 8000, 10000, 12000 and 18000 RPM. The detailed simulation parameters are provided in Table 3, which are referred with the calibrated parameters from our previous work [30]. Fig. 8 presents the DEM geometry of rotary and stationary discs in the impact pin mill. When a particle passes through the outermost pin layer leaving the impact zone, it will not be recorded in the DEM simulation. The particle impact statistic information is recorded and logged before it leaves the tracking zone. The simulation is run by 2 seconds and takes 2.5 hours to complete with 2 cores of the Intel® Xeon® E5-2600 v4 processor.

Fig. 8, 9, 10

Fig. 9 shows the snapshot of particle velocity distribution from DEM simulation and the higher velocity is observed in the outwards layers of discs. Fig. 10 shows the impact statistics predicted by DEM simulations of an impact pin mill. The mill operational speed condition has a significant effect on the impact number (average of total impacts experienced by each particle) and impact velocity. Both impact number and impact velocity increase when the pin rotational speed increases. Furthermore, there exists a marked discrepancy between the experimentally tested impact numbers, compared to the impact frequency as high as 620 calculated from DEM simulation of high rotational speed impact mill. The relationship of impact number and impact velocity associated with the rotary speed is empirically established with polynomial fitting method based on the DEM statistics. This allows to predict the average impact number per particle by pushing the boundary of rotary speed from DEM. As a result, the minimum rotary speed in this study is set as 4000 RPM whilst the maximum rotary speed is maintained as 18000 RPM as the operational limit. As seen in Fig. 10, the impact number increases from 582 until 612 with the rotary speed increases from 4000 RPM to 12000 RPM, and then the fitting curve is flattened with slight growth until 18000 RPM. In our previous study [4], the sensitivity analysis of impact number in an impact pin mill has been carried out based on population balance model where Vogel and Peukert model was adopted as the selection function. In that

work, the population balance model was used to predict the same device of impact pin mill under varying feed rates and constant rotary speed. A linear relationship between feed rate and breakage rate was built to account for the fill level. However, the sensitivity analysis of impact number was performed with a rough assumption that the impact number and impact velocity are not associated with the rotary speed.

5.2 Population balance model validation of impact number

The application of population balance model (PBM) into mineral engineering for coal particle size reduction dates back to the early work in 1950s [32]. The logic of PBM is to develop a matrix equation to track the size-mass balance evolution for every specific size range. The initial particle size distributions are described by vectors and the alterations to the size distributions resulting from the breakage process are expressed by matrices multiplying the vectors [32]. The breakage of a particle in the context of population balance model can be divided as two stages. The first stage is the selection function governing the breakage percentage of particles due to the considerable impact between particles and milling machine. At the first stage, the selection function serves to discover how particles are broken subject to the milling operation. The second stage is the breakage function to formulate broken particles in a series of size intervals, which are described by a breakage matrix. Since then, the population balance model has been extensively used to predict the ore particles size reduction in the mineral engineering [33] and many other fields such as crystallization and granulation [34–37].

For a batch milling process with well-mixed condition, the population balance model gives

$$\frac{dm_i(t)}{dt} = -S_i m_i(t) + \sum_{j=1}^{i-1} b_{i,j} S_j m_j(t) \quad (4)$$

where $m_i(t)$ is the mass of granules in the size interval i at time t . S_i and S_j are the selection function describing the fraction of particles in the size interval i and j . $b_{i,j}$ is the breakage function to describe the proportion of particles falling into j grade from grade i .

Further to Eq. (4), the selection function S_i , is the fraction by mass of particles at size interval i which are broken at time t . A conventional way for size class is to define 1 as the coarsest particles whereas size interval N is defined as the finest [38]. The selection function S_i is hereby an array with N number of elements value, describing the specific breakage rate for every size interval [39]. It can be expressed

$$S_i = S_{br,i} * P_i \quad (5)$$

where $S_{br,i}$ is the breakage rate and P_i is the breakage probability shown in Eq. (3).

The focal point of impact number n in the present work can be precisely measured by means of DEM but is conventionally obscure in the literature study of population balance model.

The breakage function in the present study is proposed by Vogel and Peukert [40] and it gives:

$$B_M = \left(\frac{x}{y}\right)^q \frac{1}{2} \left(1 + \tanh\left(\frac{y - y'}{y'}\right)\right) \quad (6)$$

where x denotes the initial particle size; y and y' denote the fragment size and the minimal particle size achieved by a milling process; q is the fitting power exponent.

Fig. 11

The procedure of validating the Vogel and Peukert model in the context of population balance model has been detailed elsewhere [4,5] and will not be expanded herein. In our previous study [4], the population balance model in Eq. (4) has been successfully applied in the impact pin mill with varying feed rates 9, 14, 19 and 24 kg/h under the constant rotary speed 10000 RPM. With a proper model calibration and validation, the parametric study of impact number indicates that lower impact velocity with higher impact numbers can give rise to equivalent consequence as higher impact velocity with lower impact numbers. The previous one is focused on the varying feed rate with constant rotary speed and merely explores PBM without the particle dynamics from DEM, which leads to the rough input for the kinetic parameters in the Vogel and Peukert model. The influence of impact number is only quantified by single variate analysis without interplay associated with the rotary speed and impact energy. As a result, the particle dynamics, i.e. impact velocity and impact number from DEM lend its support to population balance model in a refined form to examine the influence of impact number on the impact pin mill. The DEM-PBM coupling framework shown in Fig. 11 was developed to synergize the breakage kernel in the PBM with the particle dynamics from DEM [30]. In this scenario, DEM simulation of the impact pin mill serves to provide the particle dynamics, which will be directly used in the selection function of PBM. The particle size distribution information is not fed back to DEM once the PBM simulation is completed. Due to one-way coupling scenario, the arrow from PBM back to DEM is depicted in a dashed line. To take into account the size reduction in DEM simulation, the two-way DEM-PBM coupling approach needs to be implemented. This requires the updated particle size distribution from PBM to be considered in DEM for iteration which is beyond the scope of this study. In this study, the sensitivity analysis of rotary speed in a wider spectrum of rotary speed is conducted and hence the variation of impact energy and impact number is considered with the empirical relationship with rotary speed. Global system analysis (GSA) is a prominent tool to systematically explore the relative significance of model input parameters with their influences on the model output. GSA becomes increasingly useful in design space expansion and decision-making processes when the particulate process is fully validated with predictive power. The basic principle of GSA lies in the sampling for the defined particulate process, i.e. assigning a wide range of individual parameter values and calculating the responsive values from the model

outputs. In this study, the rotary speed is assigned as input variable spanning from 4000 RPM to 18000 RPM with the interval as 280 RPM, resulting in 51 testing scenarios. The model output responses include the mass-based size distribution, the quantiles and the volume fraction. In the present study, the global system analysis on the validated population balance model is carried out in gPROMS. Further description of global system analysis can be found elsewhere [41]. The advantage of global system analysis is to tackle the increasing complexity of particulate comminution system and to overcome the limitation of enormous design space from experimental characterisation. The input parameters of Vogel and Peukert model in PBM are summarized in Table 4. Note that the input values of f_{Mat} and $W_{m,min}$ are specific to the particle size range 1.4–1.7 mm, which were fitted from the single zeolite particle impact tests. Properties of particle have a significant effect on the model parameters. The relationship can be expressed as:

$$f_{Mat} = c_1 \rho \left(\frac{H}{K_c} \right)^{2.5} \quad (7)$$

where c_1 is a proportionality constant and ρ is the particle density. H and K_c are the particle hardness and fracture toughness, which are usually measured by nano-indentation test.

$$x * W_{m,min} = c_2 \frac{1}{\rho} \left(\frac{f_{Mat}}{\rho} \right)^{-1.5} \quad (8)$$

where c_2 is a proportionality constant.

The determination of $x * W_{m,min}$ is found to be increasingly inaccurate for small values of the energy threshold and exhibits larger scatter than f_{Mat} from milling experiment [13]. Moreover, this model has weakness at small breakage probability, which in turn affects the results of $x * W_{m,min}$.

The calibrated parameters are referred from the work of Wang et al. [42] and the parameters for global system analysis are referred from the DEM simulation. In this study, the DEM simulation of impact pin mill is carried out in the commercial software EDEM [43] and the PBM simulation is conducted in the advanced process modelling platform gPROMS [44]. Currently there are other different tools which allow bounded DEM particles to break during the process and this would provide alternative insights into the particle impact breakage. For example, fragmentation of bounded DEM particles is directly investigated subject to ball mill, where the reduced particle size distribution is calculated via a density-based clustering method [45].

In Fig. 12, the impact number is varied from 484 to 621 through polynomial fitting function based on the rotary speed, which totals 51 sampling points for its sensitivity analysis on the particle size distribution. The rotary speed is varied from 4000 RPM to 18000 RPM whilst impact velocity is varied from 21 m/s to 87 m/s correspondingly. The milling test is set as 20 seconds whilst the feed rate is kept constant as 24 kg/h.

Fig. 12

Given the preceding parametric study of impact number 350 in the exemplar study of impact pin mill [4], this implies that the effect of impact number will be less dominant subject to a high impact velocity. However, the sensitivity analysis was merely conducted with single variate analysis, due to the lack of synergic effect of impact energy and impact number caused from rotary speed. With the aid of DEM capacity to interrogate the particle dynamics, the empirical relationship of impact energy and impact number associated with rotary speed can be established. Fig. 13 depicts the global system analysis of impact number on the quantiles of particle size distribution. The quantiles such as d_5 , d_{25} and d_{50} are substantially decreased with the increase of impact number.

Fig. 13, 14

Fig. 14 displays the global system analysis of impact number, synergy with impact velocity on the quantiles d_{10} , d_{50} and d_{90} . Fig. 14(a) indicates the reduction of d_{10} down to 2.5 μm with the initial value of 106 μm . Fig. 14(b) implies a substantial decrease of d_{50} from 150 μm down to 31 μm when the impact number increased to from 480 to 621. Likewise, the similar trend is observed for d_{90} in Fig. 14(c) despite a less pronounced reduction. Interestingly, a piecewise function of size reduction is observed with regard to the increase of rotary speed in Fig. 14. It infers a potential avenue to improve the milling efficiency for a desired value of d_{50} by maintaining the lower boundary value of the rotary speed.

Fig. 15 displays the volume-based product size distribution in the varying regime of rotary speed. The peak value of product particle size is sharply reduced from 150 μm to 10 μm approximately. Experimental evidence has shown that increase of impact number results in the degradation of particle strength [46]. However, the maximum impact number is usually below 20 times given the experimental limitation and it will be time-consuming to experimentally carry out the impact number at the magnitude of over one hundred. The global system analysis has exemplified its power in greatly expanding the experimental space and aid the decision making in achieving the optimal quantiles in the product size distribution.

Fig. 15

6 Conclusions

The main purpose of this paper is to validate a particle breakage model with the focal point on the impact number. The validated breakage model is Vogel and Peukert model, which is widely used in the impact loading but less studied in the application of repeated impact. The theoretical background of particle strength decay is presented and it shows the breakage probability increases under increased impact number.

The selective literature breakage database is collected to examine the validity of Vogel and Peukert model in a wide variety of particle sizes and impact loading conditions. The predicted breakage probability with fitted parameters gives rise to marked agreement with the sourced breakage database irrespective of impact velocity and particle size. A multiscale DEM-PBM coupling framework is employed to validate the

effect of impact number in an impact pin mill. The dramatic size reduction in the milling process is resulted from the synergic action of impact velocity and impact number. The successful validation of Vogel and Peukert model considering the effect of impact number demonstrates its versatility including numerous key parameters such as impact energy, particle size and property.

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Appendix

Table A1 Specification of pin layer in the rotary discs

Rotary disc	Ring radius (mm)	Pin Number	Pin spacing (mm)	Pin radius (mm)	Net spacing (mm)
R1	31	24	8.11	1.5	5.11
R3	39	40	6.12	1.5	3.12
R5	47	58	5.09	1.5	2.09
R7	55	70	4.93	1.5	1.93

Table A2 Specification of pin layer in the stationary discs

Stationary disc	Ring radius (mm)	Pin Number	Pin spacing (mm)	Pin radius (mm)	Net spacing (mm)
R2	35	32	6.87	1.5	3.87
R4	43	44	6.14	1.5	3.14

R6	51	54	5.93	1.5	2.93
R8	59	62	5.98	1.5	2.98

References

- [1] E. Petrakis, K. Komnitsas, Improved modeling of the grinding process through the combined use of matrix and population balance models, *Minerals*. 7 (2017). <https://doi.org/10.3390/min7050067>.
- [2] D.M. Weedon, A perfect mixing matrix model for ball mills, *Miner. Eng.* 14 (2001) 1225–1236. [https://doi.org/10.1016/S0892-6875\(01\)00139-X](https://doi.org/10.1016/S0892-6875(01)00139-X).
- [3] R.M. De Carvalho, L.M. Tavares, Predicting the effect of operating and design variables on breakage rates using the mechanistic ball mill model, *Miner. Eng.* 43–44 (2013) 91–101. <https://doi.org/10.1016/j.mineng.2012.09.008>.
- [4] Z.P. Li, L.G. Wang, W. Chen, X. Chen, C. Liu, D. Yang, Scale-up Procedure of Parameter Estimation in Selection and Breakage Functions for Impact pin Milling, *Adv. Powder Technol.* 31 (2020) 3507–3520.
- [5] L.G. Wang, R. Ge, X. Chen, R. Zhou, H.M. Chen, Multiscale digital twin for particle breakage in milling: From nanoindentation to population balance model, *Powder Technol.* 386 (2021) 247–261. <https://doi.org/10.1016/j.powtec.2021.03.005>.
- [6] H. Kalman, Particle breakage and attrition, *KONA Powder Part.* 18 (2000) 108–120. <https://doi.org/10.1201/b10997-20>.
- [7] H. Kalman, V. Rodnianski, M. Haim, A new method to implement comminution functions into DEM simulation of a size reduction system due to particle-wall collisions, *Granul. Matter.* 11 (2009) 253–266. <https://doi.org/10.1007/s10035-009-0140-8>.
- [8] L.G. Wang, S.U. Pradhan, C. Wassgren, D. Barrasso, D. Slade, J.D. Litster, A breakage kernel for use in population balance modelling of twin screw granulation, *Powder Technol.* 363 (2020) 525–540. <https://doi.org/10.1016/j.powtec.2020.01.024>.
- [9] R. Ge, L. Wang, Z. Zhou, DEM analysis of compression breakage of 3D printed agglomerates with different structures, *Powder Technol.* 356 (2019) 1045–1058. <https://doi.org/10.1016/j.powtec.2019.08.113>.
- [10] R. Ge, M. Ghadiri, T. Bonakdar, Q. Zheng, Z. Zhou, I. Larson, K. Hapgood, Deformation of 3D printed

- agglomerates: Multiscale experimental tests and DEM simulation, *Chem. Eng. Sci.* 217 (2020) 115526. <https://doi.org/10.1016/j.ces.2020.115526>.
- [11] P.H. Shipway, I.M. Hutchings, Attrition of brittle spheres by fracture under compression and impact loading, *Powder Technol.* 76 (1993) 23–30. [https://doi.org/10.1016/0032-5910\(93\)80037-B](https://doi.org/10.1016/0032-5910(93)80037-B).
- [12] M. Ghadiri, Z. Zhang, Impact attrition of particulate solids. Part 1: A theoretical model of chipping, *Chem. Eng. Sci.* 57 (2002) 3659–3669. [https://doi.org/10.1016/S0009-2509\(02\)00240-3](https://doi.org/10.1016/S0009-2509(02)00240-3).
- [13] M. Meier, E. John, D. Wieckhusen, W. Wirth, W. Peukert, Influence of mechanical properties on impact fracture: Prediction of the milling behaviour of pharmaceutical powders by nanoindentation, *Powder Technol.* 188 (2009) 301–313. <https://doi.org/http://dx.doi.org/10.1016/j.powtec.2008.05.009>.
- [14] L.G. Austin, P. Bagga, An analysis of fine dry grinding in ball mills, *Powder Technol.* 28 (1981) 83–90. [https://doi.org/10.1016/0032-5910\(81\)87014-3](https://doi.org/10.1016/0032-5910(81)87014-3).
- [15] A.G. Evans, T.R. Wilshaw, Quasi-static solid particle damage in brittle solids -- I. Observations, analysis and implications, *Acta Metall.* 24 (1976) 939–956. [https://doi.org/doi.org/10.1016/0001-6160\(76\)90042-0](https://doi.org/doi.org/10.1016/0001-6160(76)90042-0).
- [16] A.G. Evans, M.E. Gulden, M. Rosenblatt, Impact Damage in Brittle Materials in the Elastic-Plastic Response Regime, *Proc. R. Soc. A Math. Phys. Eng. Sci.* 361 (1978) 343–365. <https://doi.org/10.1098/rspa.1978.0106>.
- [17] L. Vogel, W. Peukert, Breakage behaviour of different materials - Construction of a mastercurve for the breakage probability, *Powder Technol.* 129 (2003) 101–110. [https://doi.org/10.1016/S0032-5910\(02\)00217-6](https://doi.org/10.1016/S0032-5910(02)00217-6).
- [18] L.M. Tavares, R.P. King, Single-particle fracture under impact loading, *Int. J. Miner. Process.* 54 (1998) 1–28. [https://doi.org/10.1016/S0301-7516\(98\)00005-2](https://doi.org/10.1016/S0301-7516(98)00005-2).
- [19] A.M.E. Rizk, H.A.A. El-Sageer, M.A. Doheim, Examination of single and repetitive impact breakage, *Miner. Eng.* 7 (1994) 479–490. [https://doi.org/10.1016/0892-6875\(94\)90160-0](https://doi.org/10.1016/0892-6875(94)90160-0).
- [20] F. Shi, T. Kojovic, Validation of a model for impact breakage incorporating particle size effect, *Int. J. Miner. Process.* 82 (2007) 156–163. <https://doi.org/10.1016/j.minpro.2006.09.006>.
- [21] L.M. Tavares, R.P. King, Modeling of particle fracture by repeated impacts using continuum damage mechanics, *Powder Technol.* 123 (2002) 138–146. [https://doi.org/10.1016/S0032-5910\(01\)00438-7](https://doi.org/10.1016/S0032-5910(01)00438-7).
- [22] L.M. Tavares, Analysis of particle fracture by repeated stressing as damage accumulation, *Powder*

- Technol. 190 (2009) 327–339. <https://doi.org/10.1016/j.powtec.2008.08.011>.
- [23] M. Bwalya, N. Chimwani, Development of a More Descriptive Particle Breakage Probability Model, 10 (2020) 710.
- [24] H. Rumpf, Physical aspects of comminution and new formulation of a law of comminution, Powder Technol. 7 (1973) 145–159. [https://doi.org/10.1016/0032-5910\(73\)80021-X](https://doi.org/10.1016/0032-5910(73)80021-X).
- [25] R. Weichert, Theoretical Prediction of Energy Consumption and Particle Size Distribution in grinding and drilling of brittle materials, Part. Part. Syst. Charact. 8 (1991) 55–62.
- [26] L.G. Wang, Particle Breakage Mechanics in Milling Operation, University of Edinburgh, 2016.
- [27] E.W. Andrews, K.S. Kim, Threshold conditions for dynamic fragmentation of glass particles, Mech. Mater. 31 (1999) 689–703. [https://doi.org/10.1016/S0167-6636\(99\)00024-1](https://doi.org/10.1016/S0167-6636(99)00024-1).
- [28] Y. Rozenblat, E. Grant, A. Levy, H. Kalman, J. Tomas, Selection and breakage functions of particles under impact loads, Chem. Eng. Sci. 71 (2012) 56–66. <https://doi.org/10.1016/j.ces.2011.12.012>.
- [29] Y. Petukhov, H. Kalman, Empirical breakage ratio of particles due to impact, Powder Technol. 143–144 (2004) 160–169. <https://doi.org/10.1016/j.powtec.2004.04.009>.
- [30] X. Chen, L.G. Wang, J.Y. Ooi, A DEM-PBM multiscale coupling approach for the prediction of an impact pin mill, Powder Technol. 366 (2020) 408–419.
- [31] L.G. Wang, R. Ge, X. Chen, Establishing an oblique impact breakage master curve using a DEM bonded contact model, Comput. Geotech. 145 (2022) 104668. <https://doi.org/10.1016/j.compgeo.2022.104668>.
- [32] S.R. Broadbent, T.G. Callcott, A matrix analysis of process involving particle assemblies, Philos. Trans. R. Soc. London. Ser. A, Math. Phys. Sci. 249 (1956) 99–123.
- [33] L.G. Austin, P.T. Luckie, Methods for determination of breakage distribution parameters, Powder Technol. 5 (1971) 215–222. [https://doi.org/10.1016/0032-5910\(72\)80022-6](https://doi.org/10.1016/0032-5910(72)80022-6).
- [34] A.D. Randolph, M.A. Larson, Theory of particulate processes; analysis and techniques of continuous crystallization, Academic Press, 1971. <http://www.sciencedirect.com/science/book/9780125796507> (accessed June 14, 2017).
- [35] J.G. Osorio, R. Sayin, A. V. Kalbag, J.D. Litster, L. Martinez-Marcos, D.A. Lamprou, G.W. Halbert, Scaling of continuous twin screw wet granulation, AIChE J. 63 (2017) 921–932.

<https://doi.org/10.1002/aic.15459>.

- [36] A. El Hagrasy, L.G. Wang, J. Litster, Continuous Wet Granulation, in: Z. Nagy, A. Hagrasy, Litster JD (Eds.), *Contin. Pharm. Process.*, Springer, 2020.
- [37] L.G. Wang, J.P. Morrissey, D. Barrasso, D. Slade, S. Clifford, G. Reynolds, O.J. Y., J.D. Litster, Model Driven Design for Twin Screw Granulation using Mechanistic-based Population Balance Model, *Int. J. Pharm.* 607 (2021) 120939. <https://doi.org/10.1016/j.ijpharm.2021.120939>.
- [38] E. Bilgili, J. Yepes, B. Scarlett, Formulation of a non-linear framework for population balance modeling of batch grinding: Beyond first-order kinetics, *Chem. Eng. Sci.* 61 (2006) 33–44. <https://doi.org/10.1016/j.ces.2004.11.060>.
- [39] B. Olaleye, F. Pozza, C.Y. Wu, L.X. Liu, Population balance modelling of ribbon milling with a new mass-based breakage function, *Int. J. Pharm.* 571 (2019) 118765. <https://doi.org/10.1016/j.ijpharm.2019.118765>.
- [40] L. Vogel, W. Peukert, From single particle impact behaviour to modelling of impact mills, *Chem. Eng. Sci.* 60 (2005) 5164–5176. <https://doi.org/10.1016/j.ces.2005.03.064>.
- [41] S. Kucherenko, B. Feil, N. Shah, W. Mauntz, The identification of model effective dimensions using global sensitivity analysis, *Reliab. Eng. Syst. Saf.* 96 (2011) 440–449. <https://doi.org/10.1016/j.res.2010.11.003>.
- [42] L.G. Wang, R. Ge, X. Chen, On the determination of particle impact breakage in selection function, *Particuology*. (2021). <https://doi.org/10.1016/j.partic.2021.08.003>.
- [43] EDEM, User Defined Libraries Examples, (2017).
- [44] gPROMS Formulated Products Release 1.6, 2020. <https://www.psenterprise.com/>.
- [45] A. Krok, P. Peciar, K. Coffey, K. Bryan, S. Lenihan, A combination of density-based clustering method and DEM to numerically investigate the breakage of bonded pharmaceutical granules in the ball milling process, *Particuology*. 58 (2021) 153–168. <https://doi.org/10.1016/j.partic.2021.03.008>.
- [46] T. Han, Y. Petukhov, A. Levy, H. Kalman, Theoretical and experimental study of multi-impact breakage of particles, *Adv. Powder Technol.* 17 (2006) 135–157. <https://doi.org/10.1163/156855206775992328>.

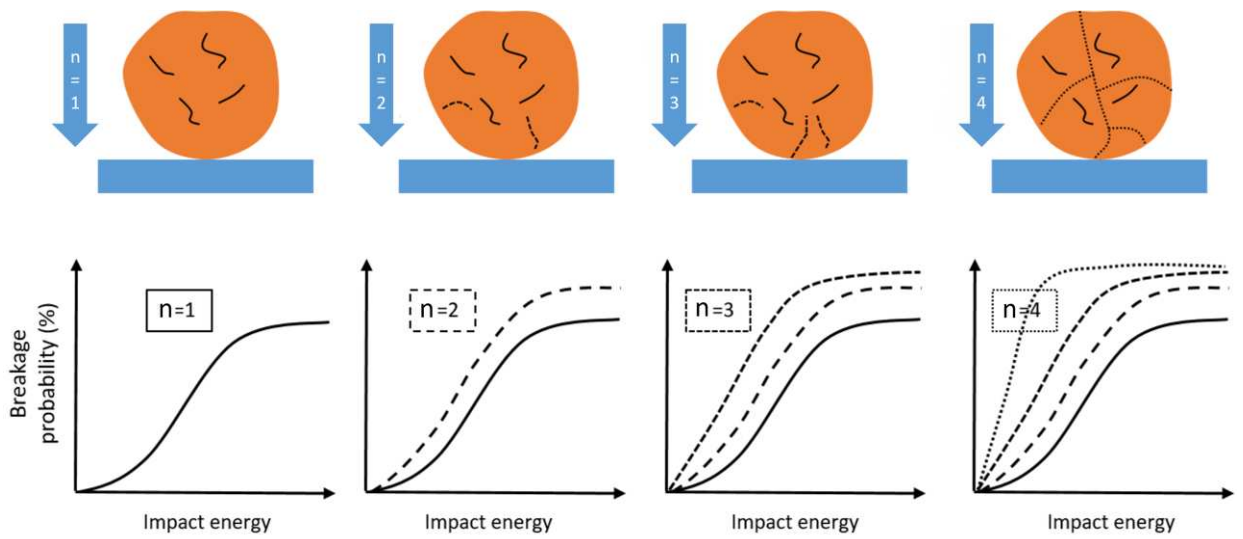


Fig. 1. Schematic of particle breakage probability by increased impact number n .

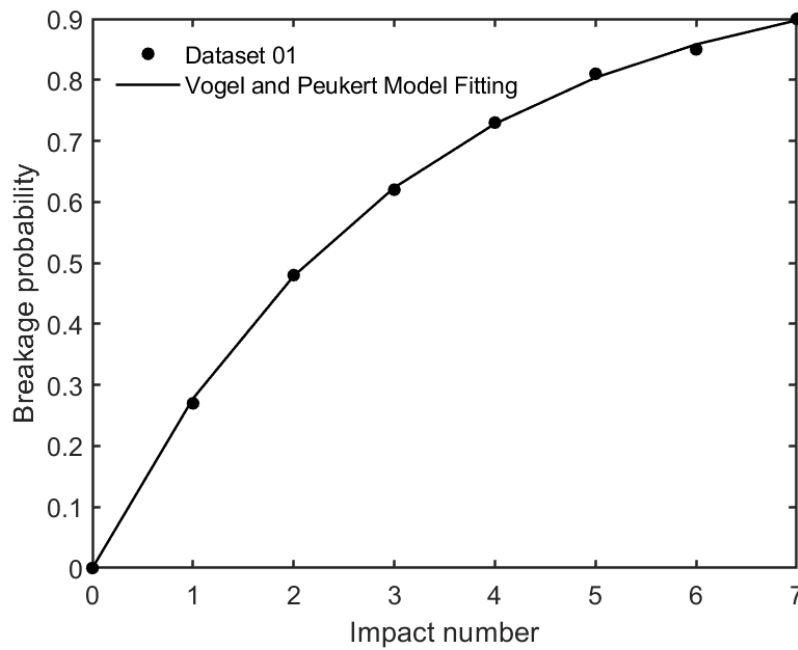


Fig. 2. Graphical comparison of breakage probability between Vogel and Peukert model fitting and experimental results from dataset 01.

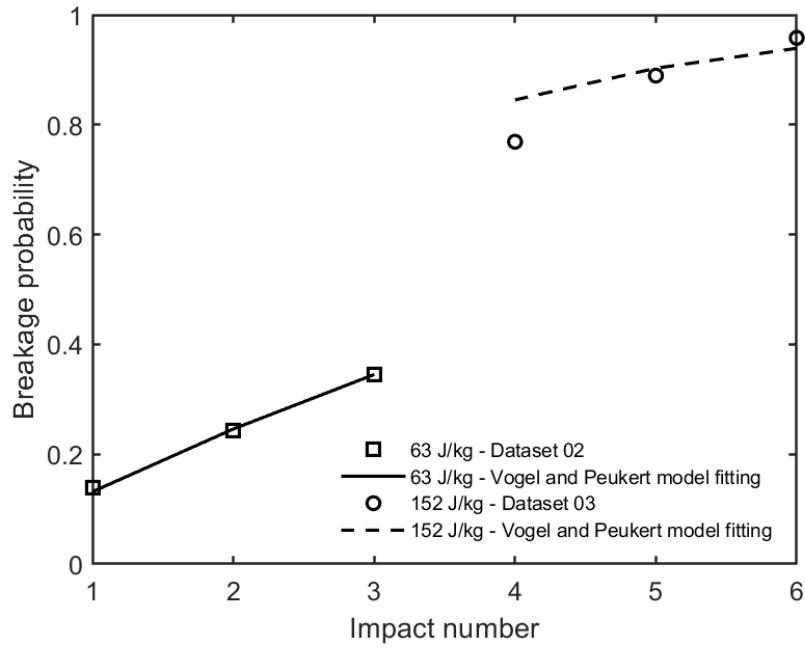


Fig. 3. Graphical comparison of breakage probability between Vogel and Peukert model fitting and experimental results from dataset 02 and 03.

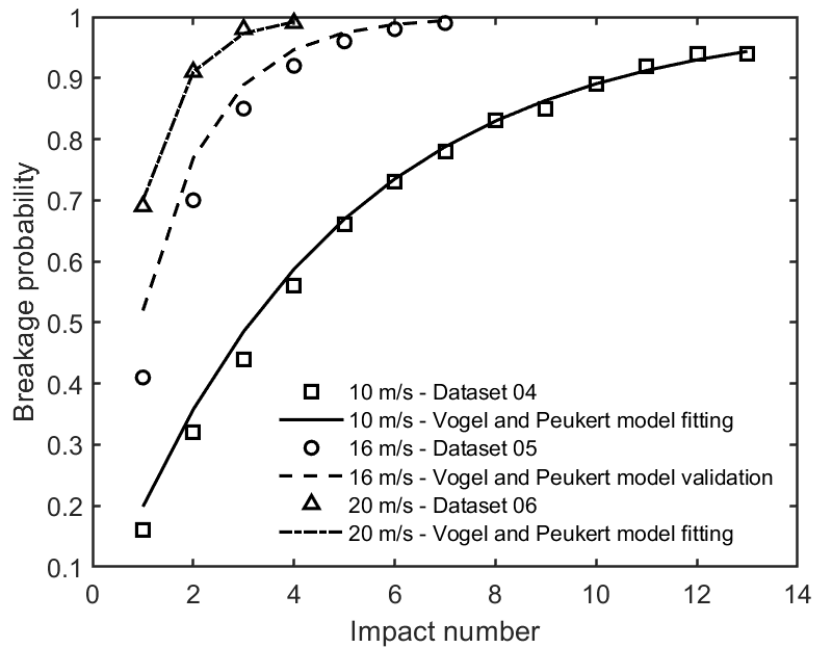


Fig. 4. Graphical comparison of breakage probability between Vogel and Peukert model validation and experimental results from dataset 04, 05 and 06.

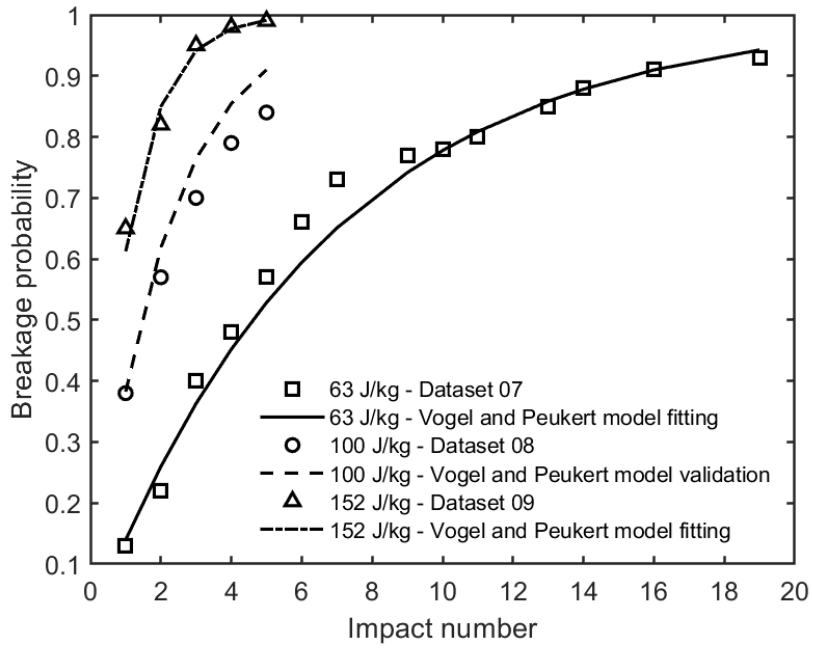


Fig. 5. Graphical comparison of breakage probability between Vogel and Peukert model fitting and experimental results from dataset 07, 08 and 09.

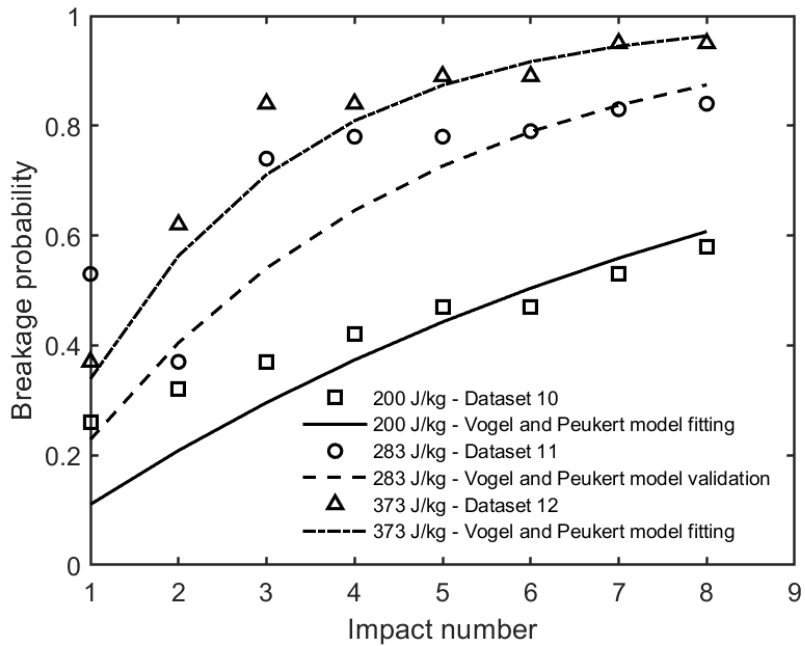


Fig. 6. Graphical comparison of breakage probability between Vogel and Peukert model fitting and experimental results from dataset 10, 11 and 12.

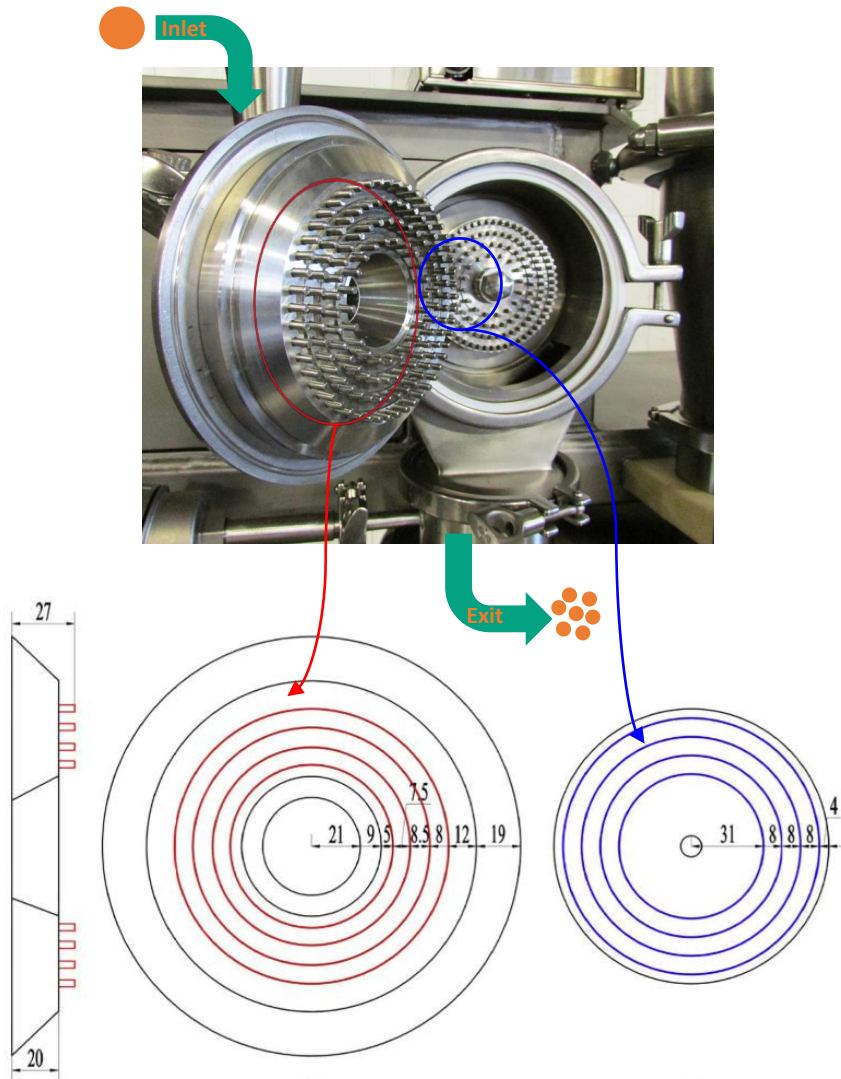


Fig. 7. Geometry of the impact pin mill in this study with red colour stationary pin layers and blue colour rotary pin layers.

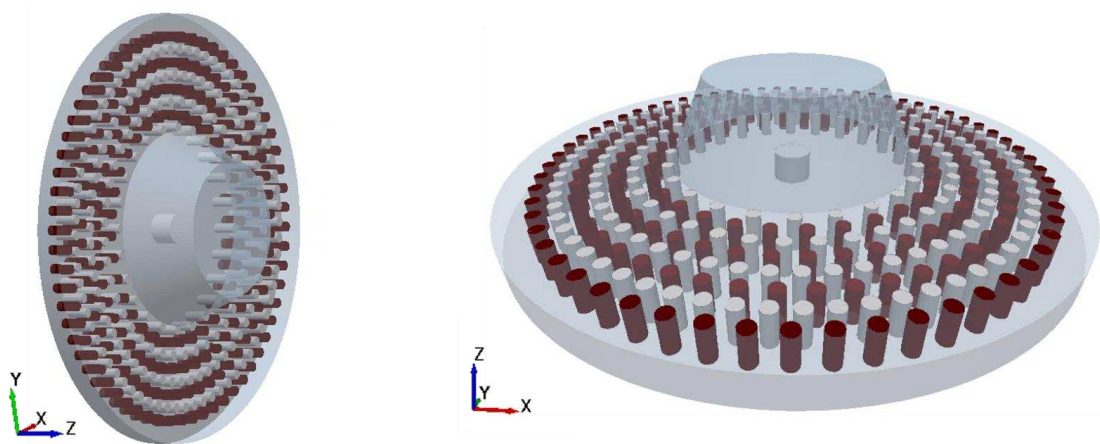


Fig. 8. Schematics of the impact pin mill used in DEM simulation.

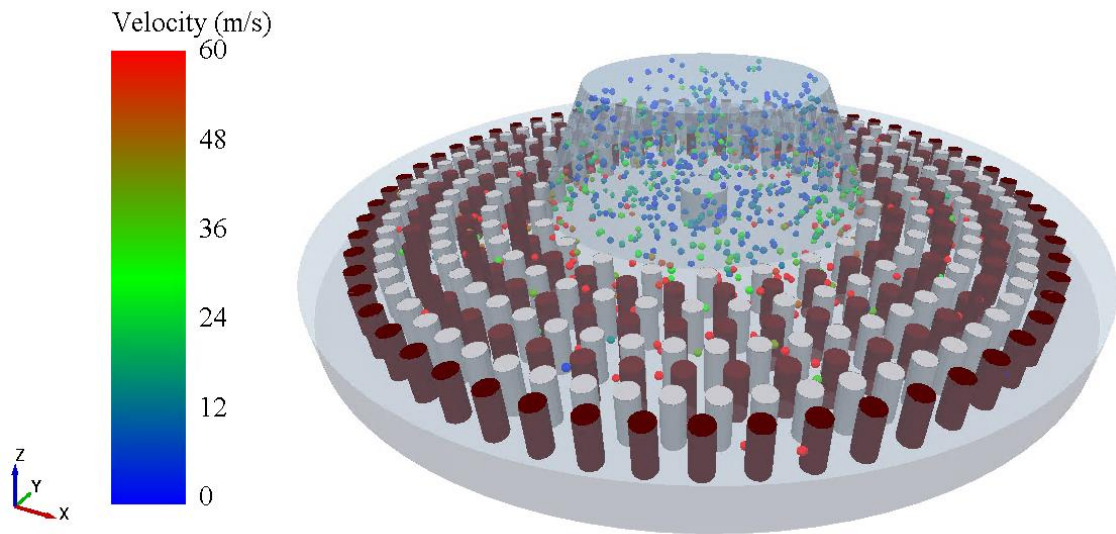


Fig. 9. DEM simulations of the particle impact in UPZ100 pin mill.

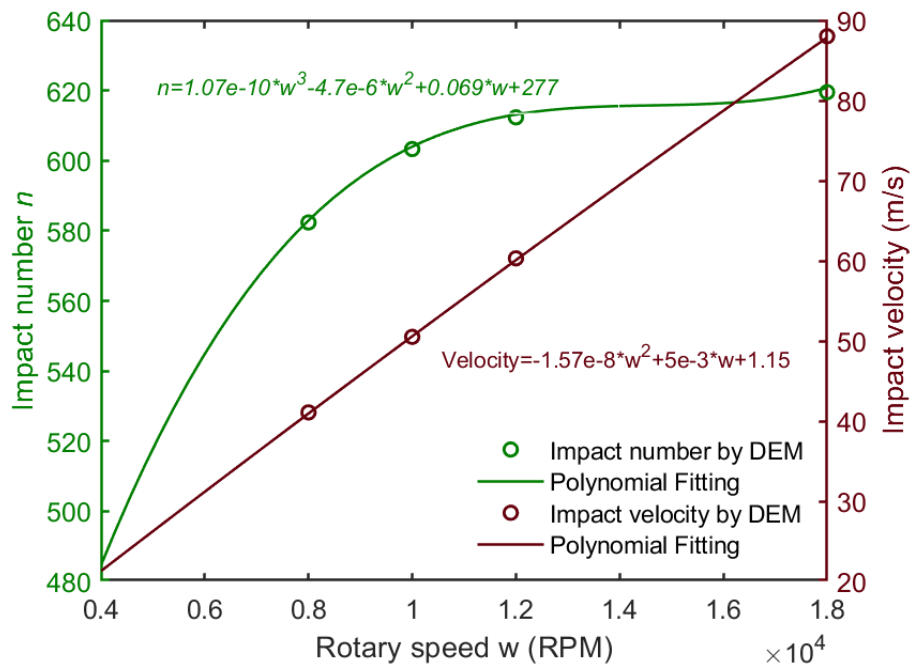


Fig. 10. The impact statistics predicted by DEM simulation of an impact pin mill. The effect of pin rotary speed on the average impact number per particle and the average impact velocity.

[Now there is only one Fig. 10, Please check (a) (b)? Thanks]

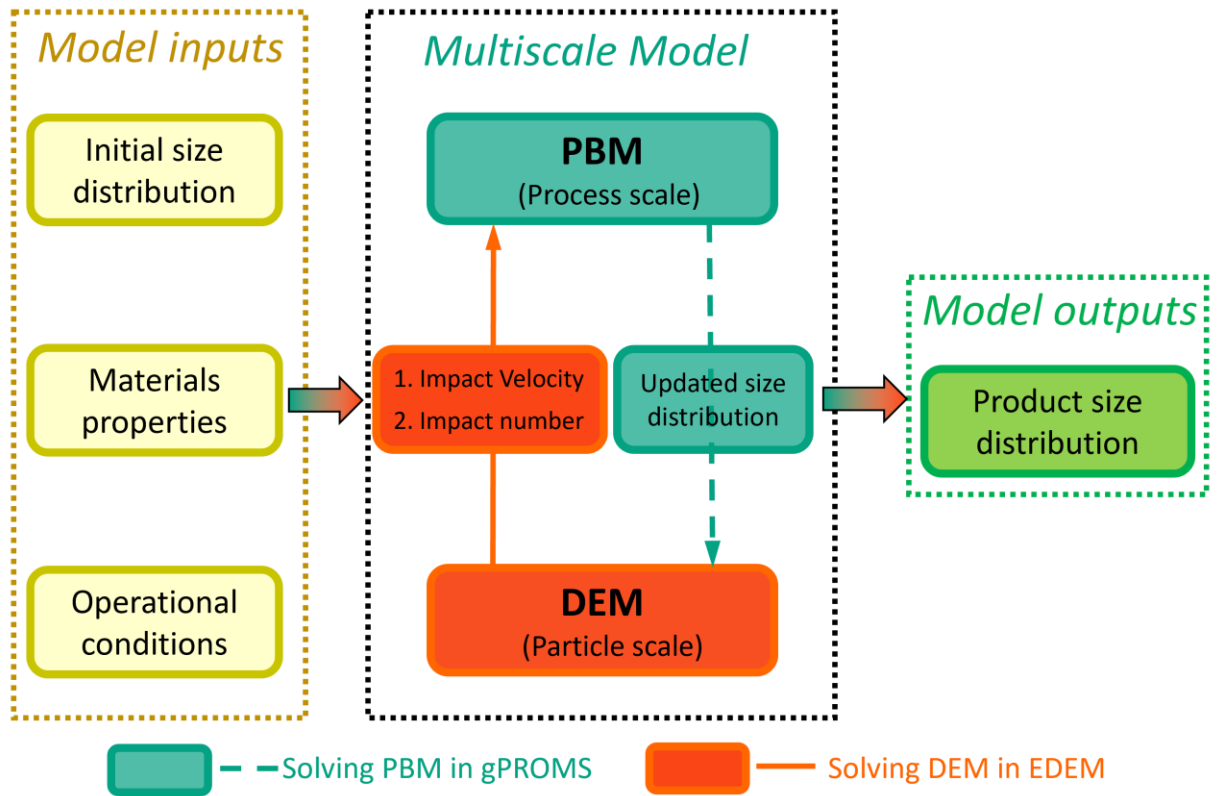


Fig. 11. DEM-PBM coupling strategy for the particle impact breakage in a milling process.

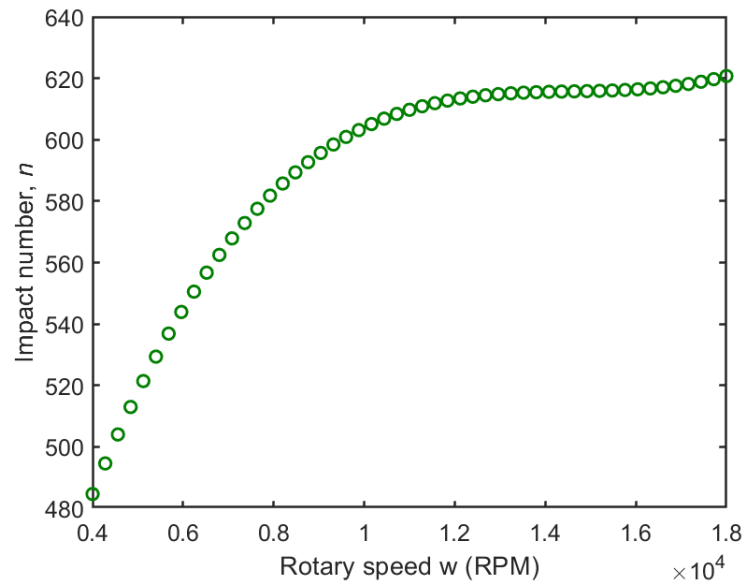


Fig. 12. The varying space of impact number as a polynomial function of rotary speed for global system analysis.

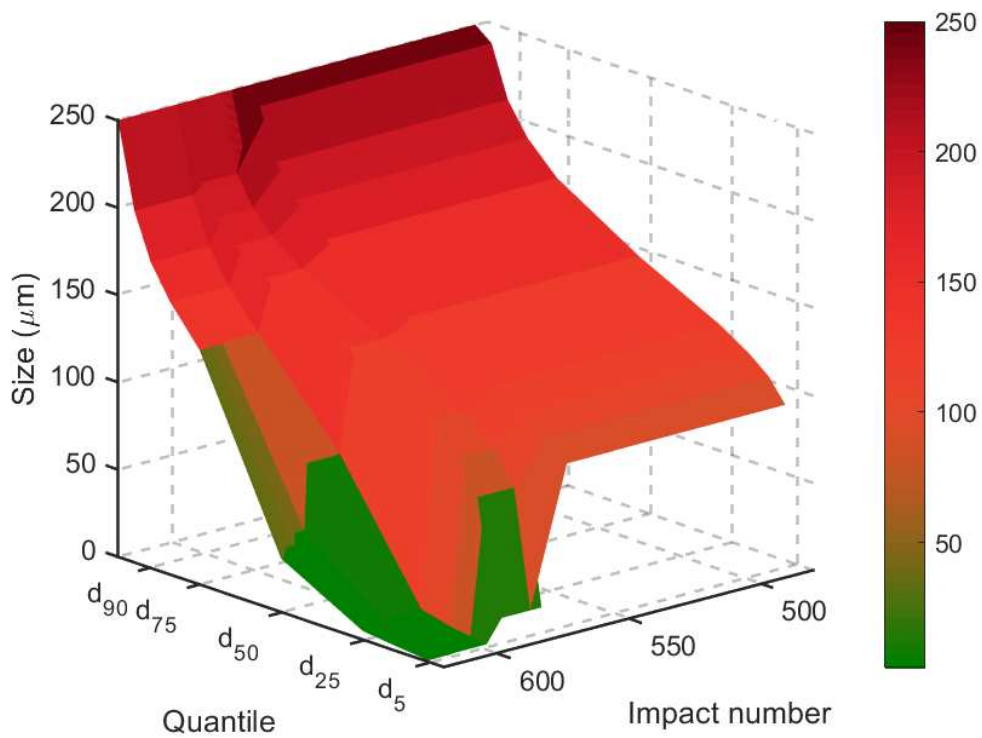
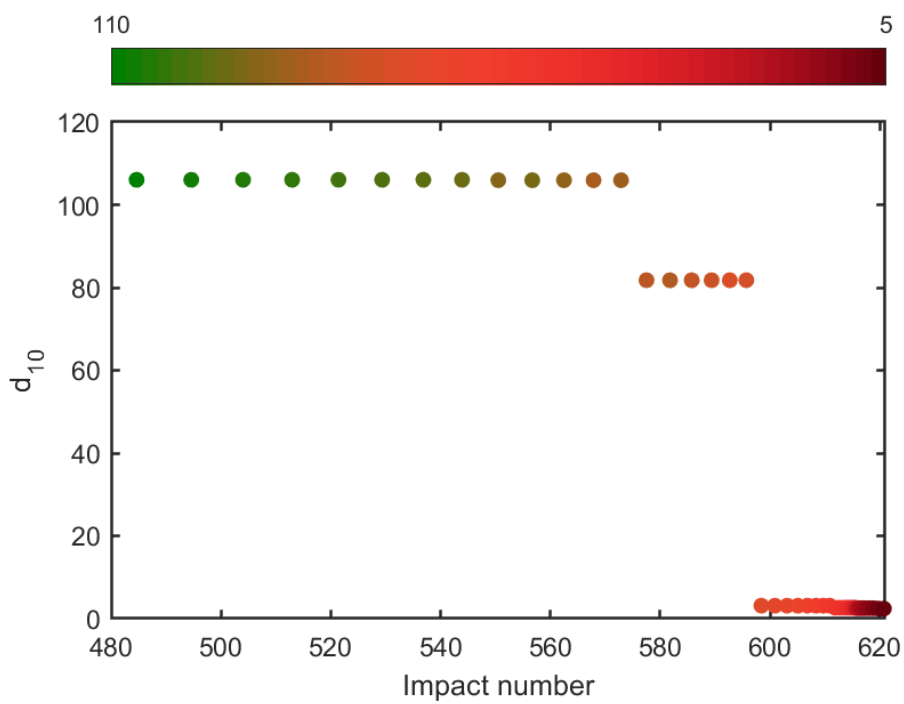
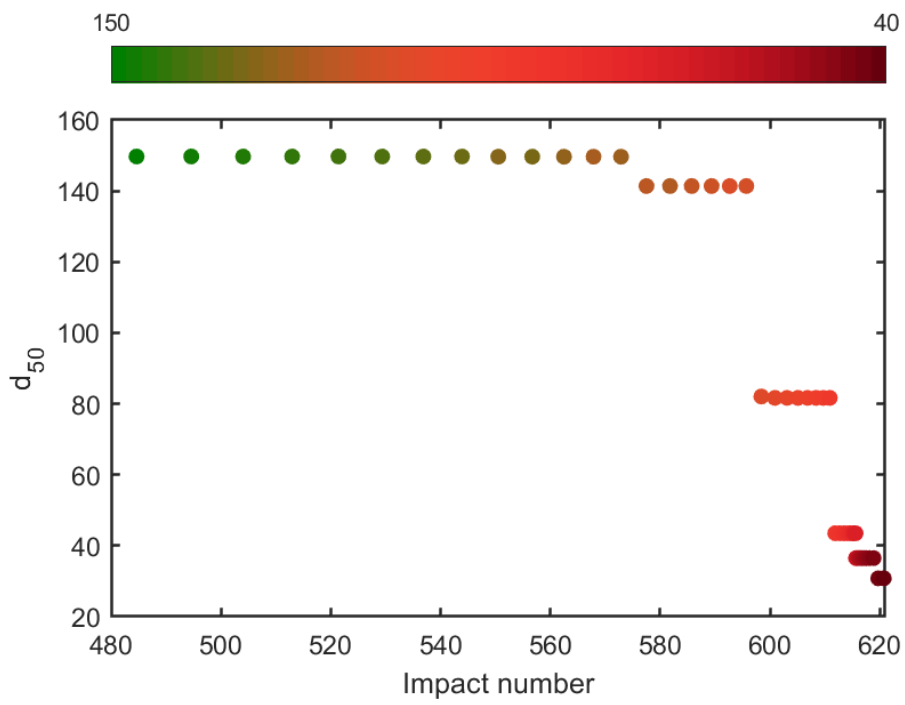


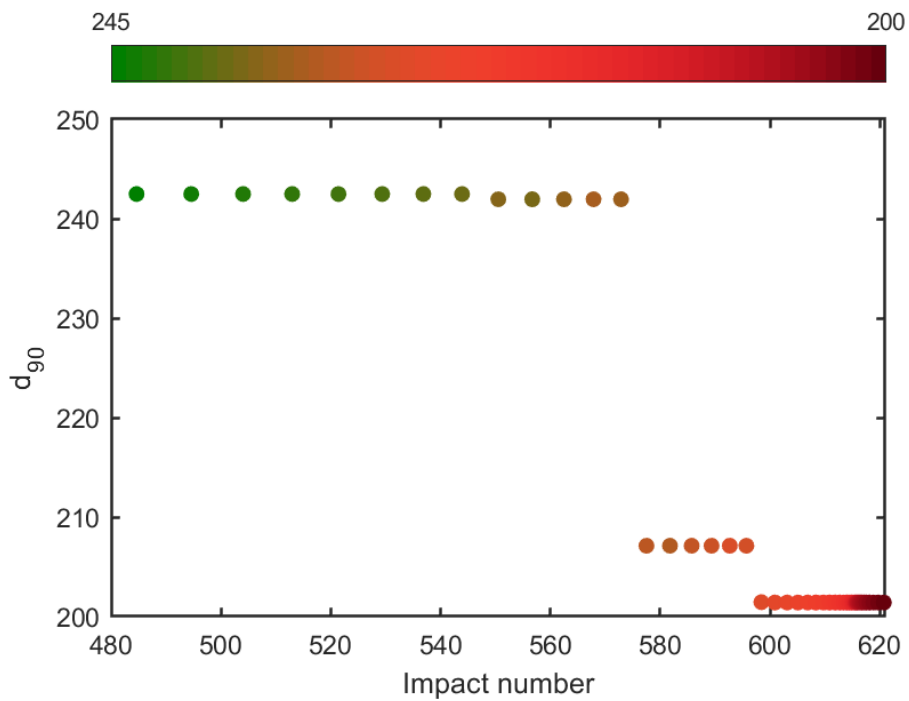
Fig. 13. Global system analysis of impact number on the quantiles of particle size distribution.



(a)



(b)



(c)

Fig. 14. Sensitivity analysis of impact number on (a) d_{10} (b) d_{50} and (c) d_{90} .

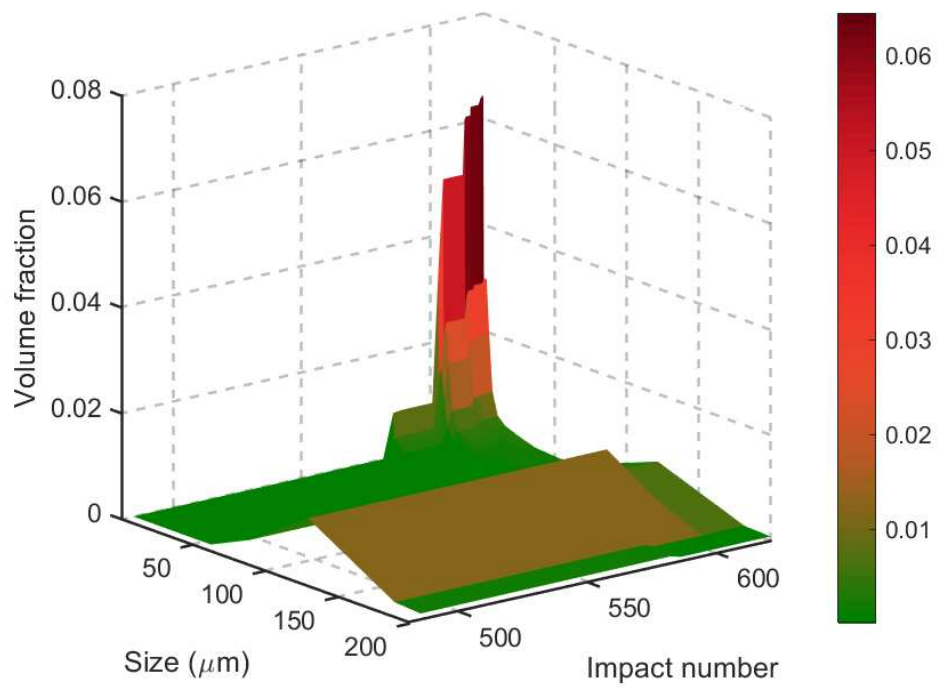


Fig. 15. Volume fraction of product size distribution under varying impact number.

Tables:

Table 1 Representative breakage database with the focus of impact number in the literature.

Dataset	Database source	Impact velocity	Impact number	Breakage probability	Tested particle	Particle size
01	Fig. 5 in Kalman et al., 2009 [7]	11 m/s	1	0.27	NaCl	1.4-2.0 mm
			2	0.48		
			3	0.62		
			4	0.73		
			5	0.81		
			6	0.85		
			7	0.90		
02	Fig. 6 in Tavares, 2009 [22]	11.23 m/s (Converted from 63 J/kg)	1	0.14	Limestone	4.0-4.75 mm
			2	0.24		
			3	0.34		
03		17.44 m/s (Converted from 152 J/kg)	4	0.77		
			5	0.89		
			6	0.96		
04	Fig. 7 in Petukhov and Kalman, 2004 [29]	10 m/s	1	0.16		
			2	0.32		
			3	0.44		
			4	0.56		
			5	0.66		
			6	0.73		
			7	0.78		
			8	0.83		
			9	0.85		
			10	0.89		
			11	0.92		
			12	0.94		
			13	0.94		
05		16 m/s	1	0.41	Dead Sea Salt	2.0-3.35 mm
			2	0.70		
			3	0.85		

		4	0.92		
		5	0.96		
		6	0.98		
		7	0.99		
06	20 m/s	1	0.69		
		2	0.91		
		3	0.98		
		4	0.99		
		1	0.13		
		2	0.22		
		3	0.40		
		4	0.48		
		5	0.57		
	11.23 m/s	6	0.66		
		7	0.73		
07	(Converted from	9	0.77		
	63 J/kg)	10	0.78		
		11	0.80		
		13	0.85		
		14	0.88		
		16	0.91	Limestone	4.0-4.75 mm
		19	0.93		
		1	0.38		
	14.14 m/s	2	0.57		
08	(Converted from 100 J/kg)	3	0.70		
		4	0.79		
		5	0.84		
	17.44 m/s	1	0.65		
09	(Converted from 152 J/kg)	2	0.82		
		3	0.95		

Fig. 8 in Tavares and King,
2002 [21]

			4	0.98		
			5	0.99		
			1	0.26		
			2	0.32		
			3	0.37		
10		20 m/s	4	0.42		
		(Converted from 200 J/kg)	5	0.47		
			6	0.47		
			7	0.53		
			8	0.58		
	Fig. 10 in		1	0.53		
	Bwalya and Chimwani,		2	0.58		
	2020		3	0.74	Gold waster rock	13.2mm
	[23]		4	0.78		
		23.8 m/s	5	0.78		
		(Converted from 283 J/kg)	6	0.79		
11			7	0.83		
			8	0.84		
			1	0.37		
		27.3 m/s	2	0.62		
12		(Converted from 373 J/kg)	3	0.84		
			4	0.84		

5	0.89
6	0.89
7	0.95
8	0.95

Table 2 Optimal values of f_{mat} and $W_{m,min}$ from different groups of dataset.

Dataset	Parameters		R^2
	f_{mat} (kg/J m)	$W_{m,min}$ (J/kg)	
01	5.57	18.75	0.999
02	0.91	24.39	0.986
03			
04	3.28	16.24	0.998
05			
06			
07	2.24	46.22	0.998
08			
09			
10	0.13	131.91	0.993
11			
12			

Table 3 DEM simulation input parameters

Parameters	Value
Particle density (kg/m ³)	2180
Particle diameter (mm)	1.6
Particle Poisson's ratio	0.3
Particle Young's modulus (GPa)	6.26
Pin density (kg/m ³)	7850
Pin Poisson's ratio	0.25
Pin Young's modulus (GPa)	81
Particle-particle restitution coefficient	0.71
Particle-particle static friction coefficient	0.35

Particle-particle rolling friction coefficient	0.11
Particle-pin restitution coefficient	0.71
Particle-pin static friction coefficient	0.13
Particle-pin rolling friction coefficient	0.11
Numerical time step (s)	3.3×10^{-7}

Table 4 Input parameters of Vogel and Peukert model in population balance model

Category	Parameters	Value	Data resource
	f_{Mat} (kg/J m)	4.64	
	$W_{m,min}$ (J/kg)	106.7	
		Lognormal distribution	Wang et al., 2021 [42]
Selection function	x	Location parameter: 1550 μm Standard deviation: 300 μm	
	$W_{m,kin}$ (J/kg)	Variable	DEM simulation
	n	Variable	
Breakage function	q	0.28	Wang et al.,
	y' (μm)	2.55	2021 [42]