



## Influence of the initial morphology on the dry-milling behavior of glass

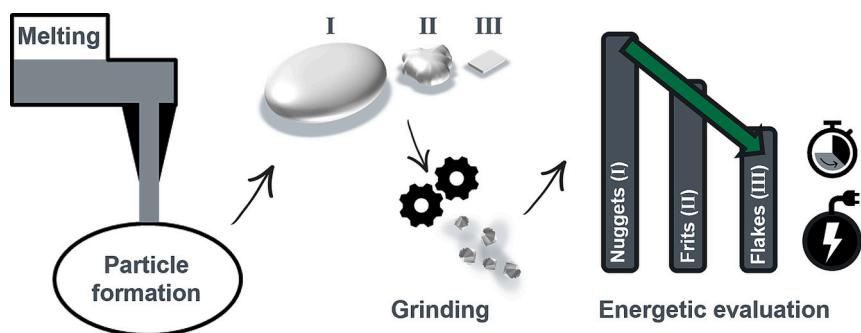
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### HIGHLIGHTS

- Evaluation of the millability of various mill feed morphologies.
- Selection of glass for forming various particles directly out of the melt.
- Identification of significant correlation between milling behavior and initial morphology.
- Substantial savings in terms of time and cost by utilization of flakes in comminution.

### GRAPHICAL ABSTRACT



### ARTICLE INFO

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### ABSTRACT

In this study, the millability of various mill feed morphologies was evaluated. Glass was selected as the material for this study due to its high degree of forming particles directly out of the melt. Our hypothesis is that an investment of additional energy in the initial processing stage of the glass will result in significant energy savings during the subsequent comminution step. The morphologies of interest were chosen as follows: namely, nuggets (I), frits (II) and flakes (III). Nuggets are conventionally cooled after cutting the melt, while frits are quenched in water, thereby permeating the chunks with cracks that facilitate their division into smaller pieces. The platelet-shaped flakes are produced by rotary atomization of the melt, and consequently, the particles exhibit a significantly smaller spatial expansion in at least one direction, which is promising for the efficient grinding of fine particles. The particle size was decreased by ball milling to a target size fraction of 63 to 250  $\mu\text{m}$ . The yield of target size fraction was evaluated, as well as the time needed to reach this particle size. A notable correlation between milling behavior and initial morphology has been identified, which may facilitate the acceleration of comminution processes.

### 1. Introduction

Key applications for finely milled glass powders include the use as functional fillers in polymers and coatings, and in the production of

fluxes, color pigments for ceramic glazes and enamels, and bioactive components in dental and biomedical materials. The particle size of the glass powder exerts a critical influence on its reactivity, processing behavior and the final material properties of the resultant product.

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Therefore, the size and size distribution must be precisely adapted to the application. In addition to the production of primary particles from glass, several researchers have also proposed the use of recycled glass as a filler material in construction materials like cement. Achieving the requisite properties of such a composite necessitates finely pulverizing the glass to a particle size of approximately 100  $\mu\text{m}$  or less [1–4]. The manufacturing of glass particles involves a series of operations, with comminution accounting for a significant portion of the electricity consumption.

In recent years, the manufacturing industry has faced increasing pressure to address environmental concerns related to energy use. The consumption of energy and its associated environmental impacts have attracted the attention of numerous researchers in this field. The glass industry, for instance, is characterized by a substantial energy demand attributed to the high temperatures necessitated by its production processes. This has resulted in a pressing need to explore diverse strategies for reducing energy consumption and, by extension, the associated carbon footprint. Notably, comminution processes are identified as a significant contributor to global energy consumption [5]. Globally, they account for an estimated 3–4 % of total electricity consumption, which corresponds to approximately 900–1200 TWh per year [6]. The comminution process utilizes a range of techniques, each meticulously designed to align with the unique characteristics of the material under consideration and the desired characteristics of the final product. The initial stage of the comminution is crushing, in which large chunks are reduced in size to form smaller, more manageable particles. Subsequently, grinding is employed to reduce the particle size to an even greater extent. The grinding process is primarily carried out by mills, which reduce the initial material to a fine powder. The reduction in particle size is achieved through the combination of abrasive and impact forces between the particles and the milling media as well as the walls of the mill. Besides this top-down approach, there are alternative methods for preparing glass powder, including bottom-up approaches like sol-gel processing [7] or gas atomization [8]. However, milling subsequent to grinding represents the predominant procedure, with the objective of achieving a further reduction in particle size. Ball mills, widely employed milling apparatus in mineral processing, are characterized by their ability to attain a substantial size reduction ratio. Despite their critical role, comminution processes are often highly inefficient with energy conversion efficiencies below 5 % [9]. Especially ball mills exhibit a relatively low level of efficiency with regard to the utilization of energy generated for the purpose of particle size reduction [9]. This highlights a significant potential for energy savings through technological improvements and process optimization. Consequently, there is a considerable need for the improvement of any of the system components to further optimize the efficiency of the mill [10]. The components of a ball mill system comprise a milling drum, milling balls, mill feed and a motor to rotate the drum.

A methodology to expedite the milling process and produce particles with a size range extending to sub-micron levels is the implementation of a wet milling process [11]. While dry milling operates without any liquid phase, in a wet mill, on the other hand, the particles are dispersed in a liquid medium during the milling process. This distinction impacts the complexity of the process, operational costs, particle size reduction ratio and the quality of the final product. The choice between wet milling process and dry milling process is influenced by factors such as particle size requirements, material sensitivity, processing time, cost, and environmental considerations. Wet milling is the preferred method for the processing of materials that are particularly susceptible to temperature-induced changes. During comminution, heat is always generated at the contact points. The liquid surrounding the particles in the wet mill acts as a heat sink, which dissipates the heat from the mill.

Another advantage is minimal yield loss. Material losses during wet milling are smaller compared to dry milling, as the wet mill is a closed system that is continuously flushed with the liquid phase. Wet milling provides finer particle sizes and better product uniformity [12]. There is also reduced dust hazard. However, wet milling tends to be more expensive due to additional equipment and processing steps. There are environmental concerns related to the wet milling process. Wet milling generally consumes a lot of water, raising environmental concerns especially in arid regions. As agglomeration has to be prevented in the case of wet milling, chemicals like dispersing agents or special solvents like xylene and toluene [13,14] as dispersing media are commonly used [15]. Consequently, wet milling can also impair the purity of the mill feed. In the case of glass, even water can be problematic. Glass materials tend to be hydrophilic, which makes drying more complex. Also, glasses with low hydrolytic or chemical resistance are not suitable for wet milling, due to the incorporation of water or leaching of components in the near-surface regions of the glass particles [16,17].

While wet milling is ideal for achieving finer sizes and managing heat-sensitive materials, dry milling is often preferred for its cost-effectiveness, greater operational flexibility, environmental friendliness due to reduced water usage, minimal waste handling, and simplicity. Thus, dry milling remains a robust and versatile option for a wide range of applications where liquid involvement is impractical. Beyond that, the size reduction range for dry milling is generally larger than for wet milling. The former is capable of processing coarser particles, while the latter typically requires a finer starting particle size for a given setup [18,19]. Therefore, a more sophisticated approach to accelerating the milling procedure would entail the examination of the influence of initial morphology on the milling process, as opposed to complicating subsequent steps of comminution by dispersing the ground material in more or less toxic media. In addition to the shape [20,21] and size of the grinding media [22,23], the performance of the mill is also known to be affected by the shape and size of the mill feed. Liu et al. demonstrated that the grinding efficiency of glass powder with a large particle size tends to decline with an increase in grinding time, approaching zero. This is due to an increase in the toughness of the glass [24]. Therefore, the optimization of the initial morphology of the grinding material represents an innovative approach with the potential to exert a significant influence on the outcome of the grinding process, particularly in the case of comminution down to the micrometer range. Given that the majority of raw materials employed in energy-intensive industries, such as ceramics and road construction, are mineral-based, the potential for pre-processing the mill feed is constrained. Conversely, the glass industry benefits from the ability to modify the morphology of glass materials by producing them directly from the melt.

A comprehensive understanding of the energy consumption characteristics of the ball mill is essential for evaluating the energy consumption and improving the energy efficiency of these grinding processes. While there are a lot of publications on modelling the influence of the technical system parameters like powder loading [23,25–27], rotational speed [23,25] or ball size [23,25,28,29], there are only a few considerations regarding the adjustment of the grinding material itself [30,31]. It is evident that glass manufacturers have significant energy requirements, which makes it imperative to explore all avenues for reducing this demand, even if it is just by reducing the energy consumption of further processing of glass to particles for the needed application [32].

The present study focuses on the production of glass particles measuring a few hundred microns through the milling process. The novelty of our work lies in evaluating the influence of initial particle morphology on the dry milling behavior of glass. The objective is to assess the impact of varying the input morphologies of the milling

process on the yield of the target fraction and potential energy savings, as well as the time required for the process. Potential pretreatment options include the targeted shaping of so-called nuggets by cutting the melt, inducing cracks through quenching the melt in water, or specifying the targeted expansion in at least one spatial direction via rotary atomization [33]. Our hypothesis put forth is that an investment of additional energy in the initial processing stage of the glass will result in significant energy savings during the subsequent comminution step.

## 2. Materials and methods

### 2.1. Materials

In the course of the tests, a modified soda-lime-silica glass was selected as mill feed due to availability reasons. The glass is compositionally similar to standard soda-lime-silica glass, with a minor addition of  $P_2O_5$ . To the best of our knowledge, this modification does not significantly alter the mechanical behavior relevant to milling. The various morphologies of the initial mill feed are compared in Fig. 1.

Glass nuggets were produced via weight-based cutting of the melt flow through a nozzle, followed by conventional cooling on a cooling belt. This process enabled the relaxation of tensions within the glass, resulting in the formation of nugget-like structures with a size of approximately 10–20 mm. Glass frits are produced identically to the nuggets, but quenched in water, thereby permeating the chunks of frits with cracks that facilitate their division into pieces of less than 1 mm. The third evaluated morphology are glass flakes, which are micron sized and platelet-shaped (flake-like) due to rotary atomization of the melt. Consequently, these particles exhibit a significantly smaller spatial expansion in at least one direction, which should be beneficial for the efficient grinding of fine particles. Glass flakes are a mass product which is commercially available and commonly used as functional filler in various applications like automotive paint [34], composite materials [35], coatings [36] and also in battery separators [37].

### 2.2. Milling procedure

A laboratory-scale ball mill with a diameter of 20 cm, a height of 24 cm, and an effective volume of  $3000\text{ cm}^3$  was utilized for the milling procedure. Two trials of dry batch milling were conducted for each mill feed, with one employing 19 mm-sized alumina ( $Al_2O_3$ ) balls and the other employing 38 mm-sized  $Al_2O_3$  balls. The ball sizes were compared to explore the influence of media size on grinding efficiency, based on the well-established principle that larger balls are more effective for coarse crushing, while smaller balls enhance fine grinding due to increased contact points [23,38]. The mill was initially loaded to approximately one-third with balls (equivalent to 2 kg), one-third with mill feed, and one-third with void space by volume. Throughout the experiment, the rotational speed of the mill was maintained at 45 rpm

(representing 50 % of the critical speed), while the residence time was varied.

The mill was operated for predetermined time intervals, with a total duration of 5, 10, 15, 20, 30, 45, 60, and 120 min, respectively. After each milling interval, the entire mill load was removed and classified by sieving. The mill load was then reloaded and milled once more until the next time interval was reached.

### 2.3. Characterization

A range of sieves with varying mesh sizes were utilized to screen the products resulting from the milling processes. This was done to ascertain the extent of size reduction (comminution ratio) and the size distribution of the product samples following each milling process. Therefore, the screening residue of particles exceeding 32, 63, 100, 250, 1000 and 4000  $\mu\text{m}$  was determined by weight. The target size fraction was set at 63 to 250  $\mu\text{m}$ , which corresponds to mesh 60 to 200. This size range aligns with common specifications for glass fillers in construction materials [3]. In order to accelerate the classification process, vibration assistance was employed for 30 min each. Additionally, the nominal particle size distribution of the glass particles in the target grinding fraction was verified by laser diffraction technology using a particle size analyzer (PSA 1190, Anton Paar, Graz, Austria). This evaluation was conducted following sieve classification to ascertain the tendency of incorrect grain entry of each morphology.

A key parameter for evaluating milling processes is the grinding rate constant, denoted as  $K_t$ . It is assumed that the sieve residue of ground material with a certain size, namely the size of the fraction coarser than the target fraction, is  $R_{\text{coarse}}$ . The value of this sieve residue is observed to gradually reduce with the prolongation of grinding time. One of the most commonly used models to describe the grinding rate is the first-order breakage rate model. It has been established from existing literature that the reduction rate of the sieve residue with a specific particle size ( $dR/dt$ ) is proportional to the sieve residue value at a given moment during the grinding process [24,39,40]. The resulting equation for this correlation is expressed as [40]:

$$dR/dt = - K_t^* R \quad (1)$$

The calculation of the grinding rate constant is conducted by using experimental data obtained from our grinding tests. As termination criterion for the time-dependent analysis a sieve residue  $R_{\text{coarse}}(t)/R_{\text{coarse}}(0)$  of less than 0.01 or a milling time of 120 min is defined.

The initial visual documentation of the source material was conducted using a stereomicroscope (Stemi 508, Zeiss, Jena, Germany). The scanning electron microscopy (SEM) imaging presented in this study was conducted using a Zeiss scanning electron microscope (LEO 1530, Zeiss, Jena, Germany) at an accelerating voltage of 5 kV. The specific surface area of the particle fractions was quantified by  $N_2$  physisorption (ASAP 2010, Micromeritics, Unterschleißheim, Germany) in accordance

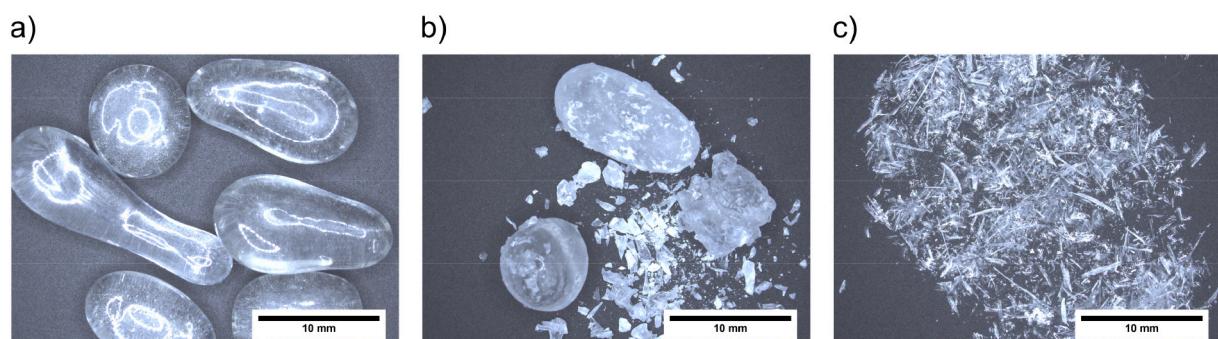


Fig. 1. Macroscopic comparison of the initial mill feed morphologies: a) Nuggets; b) Frits; c) Flakes.

with the Brunauer-Emmet-Teller (BET) equation.

To evaluate the energy consumption of a ball mill, it is necessary to measure the input power, the operating time and the mass of the mill feed or the amount of target fraction. Subsequently, the energy consumption is calculated by multiplying the power input by the operating time. The specific energy consumption per kilogram is determined by dividing the energy consumption per run by the mass of mill feed or the amount of target fraction, respectively. To draw a comparison between the mill runs, it is assumed that the power input is constant because of the constant rotational speed of the mill drum. As a result, the operating time required to produce a specified amount of target material is directly proportional to the specific energy consumption.

### 3. Results and discussion

#### 3.1. Effect of ball size

The size of the milling balls is known to exert an influence on the rate and extent of milling [23,25,28,29]. In the recent study, two ball sizes (19 and 38 mm) were employed for the grinding of three morphologies of feed material. To ascertain the alterations in particle size, the entire batch was subjected to sieving, and the respective weight fractions were quantified at designated time intervals. The weight proportion of milled particles passing per mesh width during sieve classification is shown in Fig. 2.

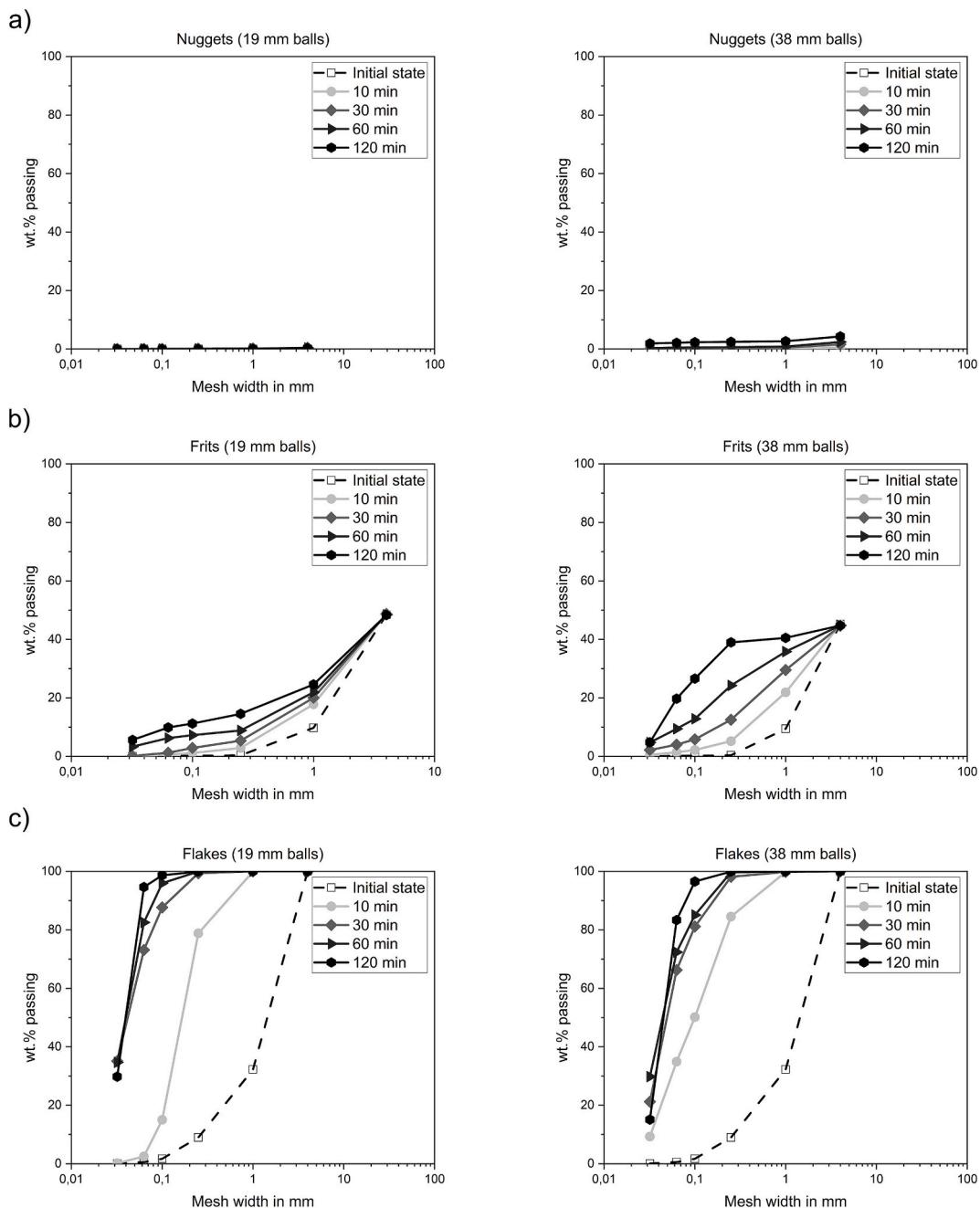
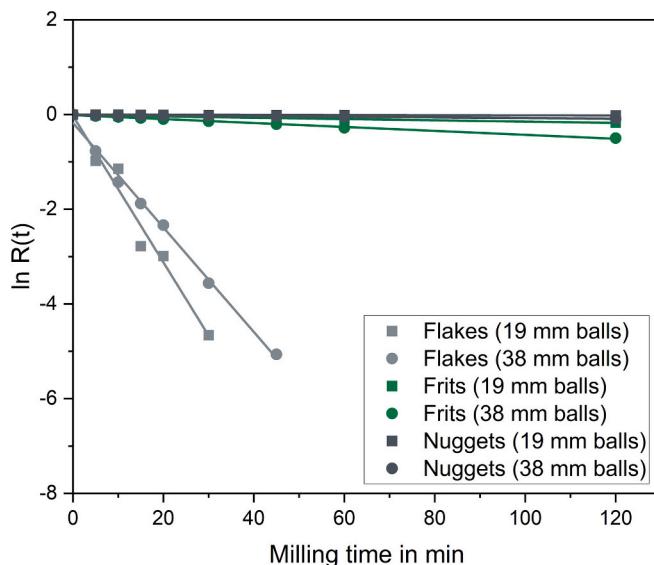


Fig. 2. Sum distribution of the particle sizes after specific milling times for the different mill feed: a) Nuggets; b) Frits; c) Flakes.



**Fig. 3.** Correlation between mass fraction of feed size and grinding time for each combination of feed morphology and ball size.

The increasing amount of weight passing the low mesh widths proves that the particles size reduces steadily with the milling time. It can be reasonably deduced that the dimensions of the milling balls employed in a ball mill have the potential to influence not only the mill's overall efficiency, but also the quality of the end product.

In the case of coarser particles at the start of the milling procedure, the application of larger milling balls results in a greater size reduction ratio than that achieved with smaller counterparts. This can be attributed to the greater kinetic energy possessed by the larger balls, enabling more effective disruption of larger particles. While the use of larger milling balls typically results in an elevated grinding rate, this may also lead to excessive wear on the mill.

For a quantitative description of the grinding kinetic behavior of glass powder, a linear regression analysis of Eq. (1) was performed on the relationships between grinding time and the logarithm of the residual mass of the coarse fraction. The fitting curves in Fig. 3 indicate a strong linear correlation from which the grinding rate constant can be determined.

In other words, the glass flakes have a significantly higher grinding efficiency than the coarser particles, which is well illustrated by the decreasing of  $K_t$  in Table 1. In this instance the bigger milling balls are less suitable. Conversely, for coarse mill feed, the larger milling balls demonstrate a higher grinding rate, which is in accordance with the observation that the ball size must be adjusted to the size of the mill feed [28,29,41].

### 3.2. Effect of morphology of mill feed on quality of milling output

In the majority of instances, the objective is not to reduce the particle size as much as possible; rather, the aim is to attain a specific target size range, as defined by the subsequent application.

For better visualization the amount of target fraction (63–250  $\mu\text{m}$ ) is plotted over time dependent on the material shape and the ball size in Fig. 4.

**Table 1**

Grinding rate constants for each combination of feed morphology and ball size.

	Flakes (19 mm balls)	Flakes (38 mm balls)	Frits (19 mm balls)	Frits (38 mm balls)	Nuggets (19 mm balls)	Nuggets (38 mm balls)
$K_t$	0.1547	0.1104	0.0014	0.0042	0.0002	0.0007
$R^2$	0.9707	0.9960	0.9814	0.9959	0.8533	0.9813

The initial particle size of nuggets and frits exceeds that of flakes as a consequence of the fabrication process. This is reflected in the high quantity of coarse fraction at the start of comminution in Fig. 4. Regardless of this fact, it is evident that the most expedient method for achieving a high target fraction is the utilization of platelet-shaped particles as mill feed. Given the elevated grinding rate, the optimal target fraction of approximately 70 wt.% is already achieved within five minutes, with only 5 wt.% of particles exhibiting a lower size in the fine fraction. Particles in the coarse fraction remain available for further milling. As the milling procedure persists, the amount of fine fraction increases, concurrently reducing the amount of target fraction for the flake material (see Fig. 4). This is due to the fact that the target fraction is not removed immediately from the system but rather remains in the ball mill where it is subjected to constant reduction in size.

While there is a lack of sufficient size reduction of nuggets to attain the target size for both ball sizes, there is a consistent increase in particles from frits within the targeted size range over time. In the case of coarse particles, such as frits, the degree of size reduction seems to be higher in relation to increasing ball size. This can be attributed to the greater kinetic energy possessed by the larger balls, enabling more effective disruption of larger particles. Also, the application of larger milling balls results in a greater amount of target fraction than that achieved with smaller counterparts. On the other hand, the use of smaller milling balls facilitates a more regulated and controlled batch-wise operation of the mill.

The particle size distribution determined by laser granulometry in contrast to the target size fraction of the classification process by sieving is shown in Fig. 5.

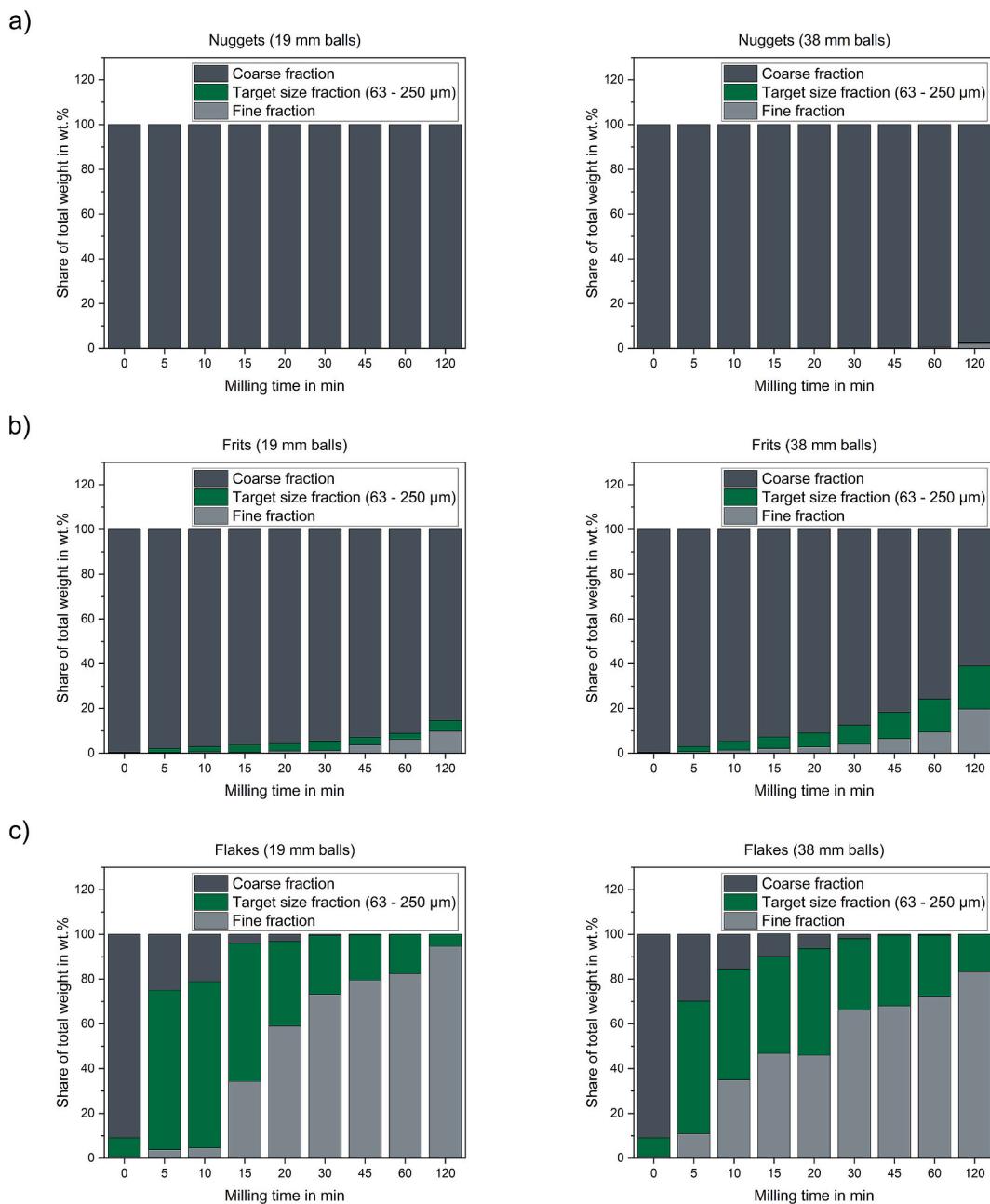
The assumed particle size range after sieving (63–250  $\mu\text{m}$ ) is best represented by the frits. It is evident that particles derived from flakes and nuggets exhibit a significant amount of incorrect grain entry during classification by sieving. In the case of platelet-shaped flakes intended as mill feed, the low expansion in at least one dimension could lead to this phenomenon [42]. In the case of chunky nuggets in the initial state, there seems to be a deviation of morphology during comminution. This prompts the question of the resulting particle shape after the milling process.

### 3.3. Effect of morphology of mill feed on resulting particle shape

The surface morphology of the mill feed in the initial state prior to comminution is depicted in the electron micrographs in Fig. 6.

From this microstructural overview, it can be observed that the surface of the glass nuggets (too large for scale) and flakes in the initial state is relatively smooth. It is evident that the frits display microcracks that originate from the quenching process during production. These already existing breaking points may facilitate size reduction at the beginning of the comminution process.

The morphology of the resulting particle shape after comminution has an impact on the potential applications of the glass particles,



**Fig. 4.** Amount of fine, target (63–250 µm) and coarse fraction after each milling interval: a) Nuggets; b) Frits; c) Flakes.

influencing a range of properties including specific surface area, bulk density, pourability and flowability. Particles of the target size fraction between 63 and 250 µm after milling for 120 min are displayed in Fig. 7.

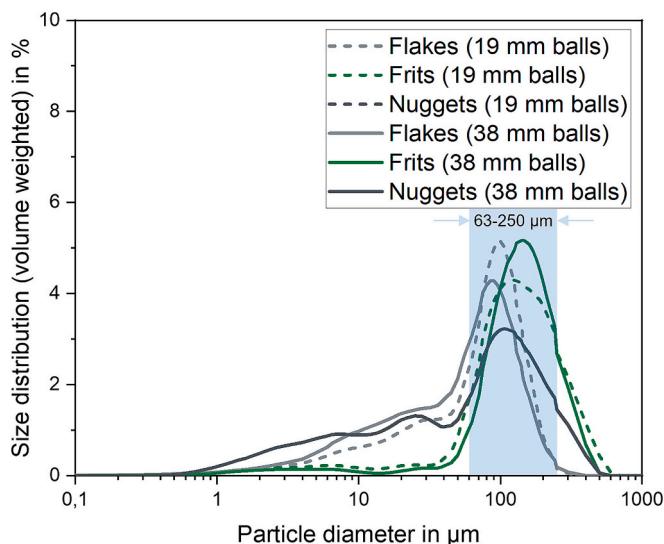
It has to be noted that no nugget-derived particles within target size range were obtained with the small milling balls. The target size fraction of nuggets milled by 38 mm milling balls and frits display a predominantly angular, block-like morphology following a milling duration of 120 min. As a consequence of abrasion, the nuggets also yield fine particles with granular and clastic morphology, which increase in number as coarse particles with angular and block morphology are worn away. This process occurs over the course of increasing milling time.

Flakes crack preferably in longitudinal direction, which results in a notable reduction in edge length while maintaining a constant thickness. This indicates that there is merely a reduction in aspect ratio, but the platelet-shape stays intact.

The equivalent particle size of glass powder is defined as a

corresponding particle size value when the cumulative percentage of the particle size distribution reaches a certain value. Thus, the equivalent particle sizes (D10, D50 and D90) of particles derived from the different mill feed morphologies are evaluated based on the assumption of spherical shapes. Furthermore, this study correlated the resulting specific surface area with the latter. Particle properties after the milling process are summarized in Table 2.

The results reveal a clear trend. After milling for 120 min, the equivalent particle size of the frit-derived glass powder is well within the target size range and its specific surface area is by far the lowest of the three morphologies. This confirms the advantages of the resulting cube-like particle shape. The powders derived from nuggets and flakes exhibit a high level of fineness, while their D50 after milling for 120 min is in the range of 58 to 75 µm. This size marks the lower end of our target size range (63 µm), indicating that the majority of particles fall below it, which is consistent with the SEM images in Fig. 7.



**Fig. 5.** Particle size distribution of the sieve-classified size fraction between 63 and 250  $\mu\text{m}$  after 120 min of milling, determined by laser granulometry.

Thus, the low specific surface area of the frit-derived particles can be attributed to the narrow particle size distribution of these particles. In contrast to ground flakes or nuggets, the proportion of fine particles is relatively low. It can be observed that an increase in the quantity of fine

particles also results in a corresponding increase in the specific surface area of the particle collective.

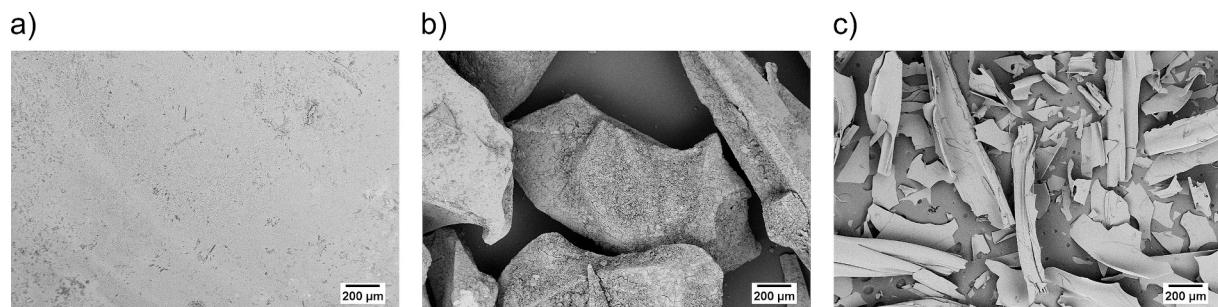
For a given product size distribution, an excessively high energy input results in products containing a high proportion of newly created particles. This indicates that a lower energy input would be sufficient for the comminution of this specific particle type. In such instances, the production of irregular particles is more pronounced. Less energy input can be achieved by either decreasing the diameter of the milling ball or reducing the rotational speed of the mill. Conversely, when the energy input is insufficient, the grinding process relies on repeated fracturing and wear action for size reduction, resulting in the production of a more rounded product particles. [43]

This batch size reduction test demonstrates that the benefits of the high millability of platelet-shaped mill feed should be exploited for

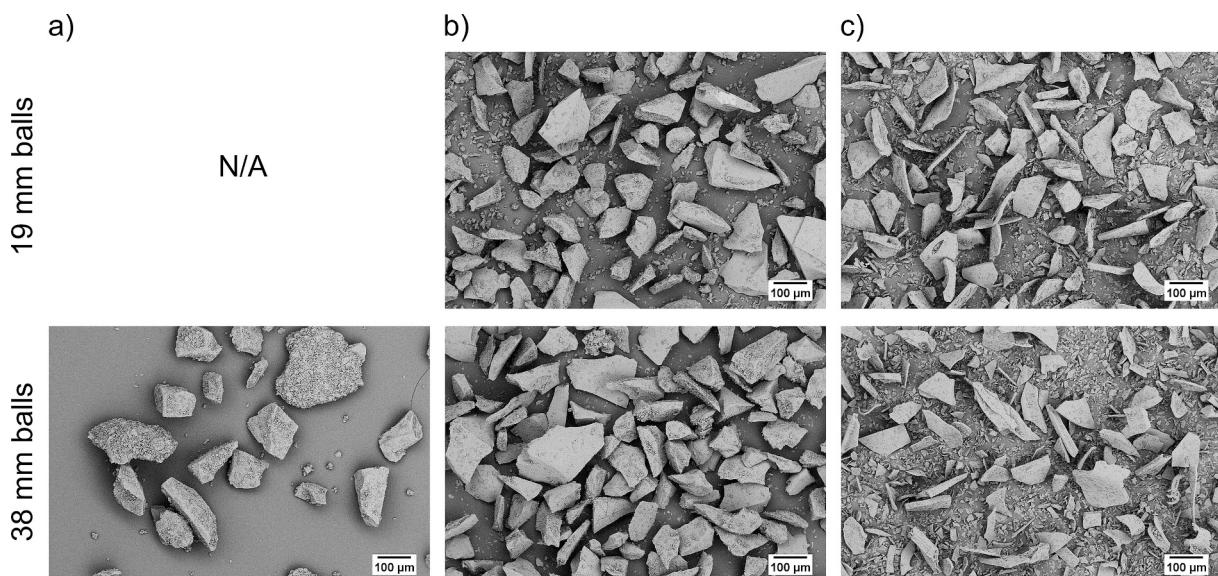
**Table 2**

Particle properties of the size fraction between 63 and 250  $\mu\text{m}$  after milling for 120 min.

Fraction 63–250 $\mu\text{m}$	Flakes 19 mm balls	Flakes 38 mm balls	Frits 19 mm balls	Frits 38 mm balls	Nuggets 19 mm balls	Nuggets 38 mm balls
Specific surface area in $\text{m}^2/\text{g}$	0.36	0.41	0.21	0.13	N/A	1.08
D10 in $\mu\text{m}$	9.96	7.34	38.30	58.20	N/A	3.92
D50 in $\mu\text{m}$	75.18	58.32	120.74	130.69	N/A	65.59
D90 in $\mu\text{m}$	141.98	131.09	263.78	239.89	N/A	196.23



**Fig. 6.** Initial surface morphology of the different mill feed prior to comminution: a) Nuggets; b) Frits; c) Flakes.



**Fig. 7.** Particle morphology of the size fraction between 63 and 250  $\mu\text{m}$  after milling for 120 min: a) Nuggets; b) Frits; c) Flakes.

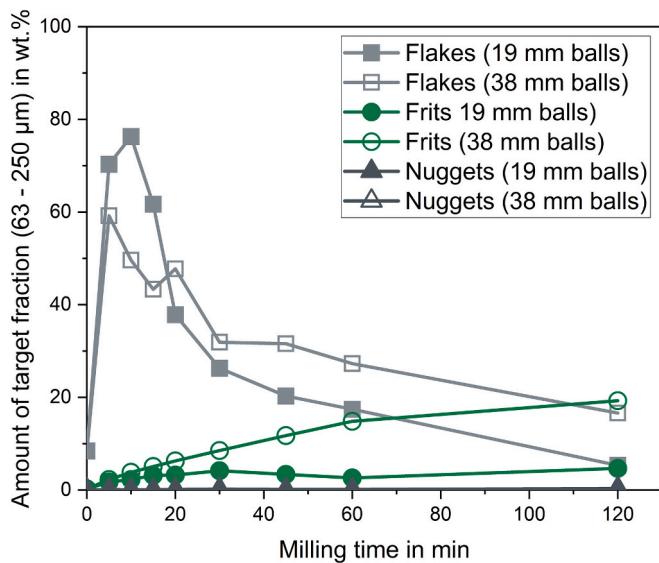


Fig. 8. Weight percentage of target size fraction at each time interval for the different mill feed and ball sizes.

applications where regular particle morphology is not necessarily required.

#### 3.4. Energetic assessment

Upon examination of the highest yield of target fraction for each morphology and ball size combination in Fig. 8, it becomes evident that platelet-shaped particles with small milling balls represent the most promising candidate for an effective and efficient milling process.

As the comminution continues, the fine fraction forms a cushion around the particles, which has the effect of slowing down the milling kinetics and finally decreases the amount of target fraction. Accordingly, for batchwise milling, the process has to be stopped before this stage is reached, because the fine fraction would increase at the expense of the target fraction. The optimum milling time can be deduced from Fig. 4 and is estimated to be between five and ten minutes for flakes, while a considerable quantity of coarse fraction still remains after 120 min when using frits or nuggets as mill feed. Those larger particles are initially crushed mainly by volume crushing, which results in high grinding efficiency. However, as grinding time increases and particle size decreases, the toughness of glass powder improves, while the volume crushing effect based on collision weakens. This results in a decrease in grinding efficiency over time. [24]

A volume of one-third of the ball mill was filled with the glass particles for each mill run. In terms of weight, this comprised 500 g of the frits and nuggets but only 250 g of the flakes due to their fluffy appearance. It is therefore possible to calculate the specific time, and, consequently, the energy consumption per kilogram of target-size particles, based on the measured power consumption of the mill and the milling duration that has been determined for the optimum yield of each mill run in Fig. 8. The results are displayed as a logarithmic plot in Fig. 9.

We assume that the energy consumption of the mill is directly proportional to the milling duration. It is evident that, despite the augmented time expenditure associated with the cleaning cycles necessitated by the diminished filling weight per batch of flakes, the high yield of the target fraction (76 and 59 wt.%, respectively) following a brief period of milling is considerably higher than that of nuggets or frits. The additional energy expenditure associated with flake production is confined to the rotation and the induction heating of the cup. Based on our previously conducted trials, an expenditure of approximately 0.5–2 kWh of electrical energy per kg of flakes can be deduced at a rotational speed of 12,000 rpm and a temperature of 1300 °C.

This means that even when the additional energy demand of the flaking process is taken into account, the utilization of flakes as starting material for comminution leads to substantial savings in terms of time and cost, by several orders of magnitude.

For a comprehensive understanding of milling efficiency, it is also essential to consider the targeted size fraction since the energy requirements for milling increase with decreasing targeted particle size [44,45]. Beyond that, it is imperative to consider any environmental factors that may potentially influence the energy efficiency of the mill. Such factors include mill size, rotating speed, the level of filling of the milling balls, the grindability of the material, as well as the size and density of the particles. However, this exceeds the scope of the present study and requires further validation through subsequent experiments.

#### 4. Conclusions

We varied the initial morphology of the feedstock for a batchwise milling process in order to minimize the milling time and optimize the yield of target fraction. The dry-milling behavior of three different glass particle morphologies was compared. Nuggets were conventionally cooled after cutting the glass melt, while frits were quenched in water, thereby permeating the chunks with cracks that facilitate their division into smaller pieces. The platelet-shaped flakes were produced by rotary atomization of the melt, and consequently, the particles exhibit a significantly smaller spatial expansion in at least one direction. Our hypothesis was that an investment of additional energy in the initial processing stage of the glass particles will result in significant energy savings during the subsequent comminution step.

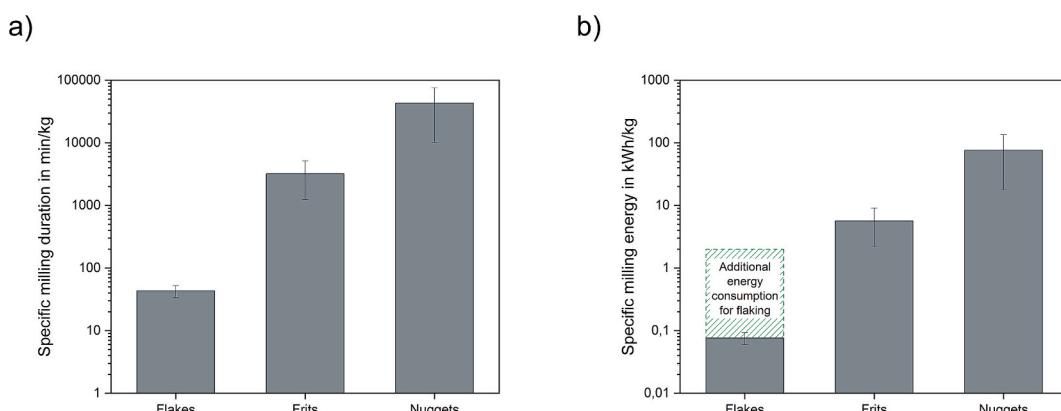


Fig. 9. Comparison of the milling behavior: a) Specific milling duration and b) energy per kg of target size fraction (63–250 μm) calculated by the yield and optimum milling time of batchwise mill runs.

The targeted fraction size for this study was 63–250  $\mu\text{m}$ . After defined milling intervals, the particle size distribution was determined by sieve classification. The peak weight fraction of milled glass flakes in this range was approximately 70 wt.% after 5–10 min and subsequently decreased as milling continued, due to the formation of a fine fraction. The highest quantity of glass frits and nuggets within the target size was attained at 120 min, which represented the conclusion of our observation period. Thus, the utilization of glass flakes as the starting material is effective as long as the morphology of the milling outcome does not have to be equilateral. While cube-like or potato-shaped particles are obtained when frits or nuggets are ground, the morphology of particles derived from the flakes remains platelet-shaped. This results in a broader particle size distribution within the sieved target fraction, as observed in the particle size distributions measured by laser granulometry.

Additionally, two sizes of milling balls were compared. It was shown that larger milling balls can effectively crush coarse particles, especially at the start of the milling process due to the higher energy input. However, as the size of the particles decreases, the smaller milling balls exhibit a higher size reduction.

This study demonstrates that at consistent operational parameters, the size and morphology of the mill feed can markedly influence the grinding rate, consequently reducing time and energy consumption of the comminution process significantly. Furthermore, the quality of the output is contingent upon the quality of the initial materials, thereby allowing for adaptations to tailor the particles to specific applications. The comminution process of glass particles was described quantitatively using a grinding kinetic equation. An enhanced grinding rate is facilitated by the utilization of platelet-shaped glass particles as the initial material source. The platelet shape inherently provides a low particle size in at least one dimension, which results in a fracture mode that is particularly favored along the edges of the particle. In contrast, glass frits have cracks in the particles as a consequence of the melt quenching process that takes place during production. This results in the easier comminution of larger chunks but has a minimal impact on smaller particles. Nuggets, which are formed from the glass melt and cooled in a conventional manner, have different milling characteristics, as they only break down by means of volume crushing.

In conclusion, when the primary objective of comminution is to achieve a specific particle size in one or two dimensions, the utilization of platelet-shaped mill feed is advantageous. Notwithstanding the consideration of the energy expenditure associated with rotary atomization processing, substantial time and thus energy savings remain. Despite the elevated comminution rate, it is essential to further optimize the milling process of the flakes with respect to duration and particle size control to ensure the minimization of comminution losses in the form of fine fraction. This can be achieved, for example, by an integrated classification unit, which is commonly used in industrial applications.

This study provides a foundation for further research on the role of particle morphology in comminution efficiency. Future work could explore a broader range of glass compositions and morphologies, including recycled and specialty glasses, to assess the generalizability of the observed trends. Additionally, incorporating advanced breakage models, such as the primary breakage distribution function as described by Austin and Luckie [46], could provide deeper insights into the fragmentation mechanisms. Scaling up the process from lab-scale to pilot or industrial levels would also be valuable for practical applications in glass recycling and powder production. Finally, coupling experimental results with numerical simulations could enhance predictive capabilities for the optimization of milling parameters.

#### CRediT authorship contribution statement

**Philipp Rank:** Writing – original draft, Visualization, Validation, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Nicolas Hacker:** Writing – original draft, Validation,

Methodology, Investigation, Formal analysis, Conceptualization.

**Thorsten Gerdes:** Writing – review & editing, Supervision, Resources.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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#### Data availability

Data will be made available on request.

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