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Comparative investigation of glass waste grinding in various mills

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Preliminary communication

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Abstract
The present work deals with systematic grinding investigation and determination of grindability of container glass bottles. The systematic grinding tests were carried out in three different kinds of ball mills (a drum mill, a vibrating mill, and a planetary mill) with different energy intensities (low, medium, and high intensity) in dry conditions. In addition, the specific grinding work and specific surface area were determined in every case. The grindability test was performed by the Universal Hardgrove Mill, moreover, the Bond-Work Index was calculated from the Hardgrove Grindability Index. In this research work, the focus was on analysing the grindability of container glass bottles and the goal was energy-efficient milling of glass waste to produce glass foam powder for further utilization (for example glass foam) in a sustainable way. Based on the obtained results, it can be concluded that the optimal milling apparatus for container glass grinding is the drum mill. The reason why is that it has a low specific grinding work (151.52 kWh/t) in comparison with the other two types of mills (3488.37 kWh/t for the planetary mill and 1106.38 kWh/t for the vibrating mill) resulting in a relatively high specific surface area (11314 cm^2/g). In the case of grinding industrial quantities of glass waste, the drum mill has a much higher capacity compared to the vibrating mill and the planetary mill.

Keywords:
glass waste; recycling; grinding; ball mill; specific grinding work

1. Introduction

Glass is an amorphous, non-crystalline solid material generally formed by rapid cooling of the molten form. The process requires a massive amount of energy. Glass is mainly used in the packaging and construction industries, so it is present in the area of household and construction waste (**Spence et al., 2016, Shelby, 2017**). Glass waste is not naturally degradable, which results in significant environmental problems if it is not dumped in a controlled manner. On the other hand, glass waste has high recycling potential because it can be fully recycled, which means that one tonne of glass waste can be converted into one tonne of new glass product by adding energy (**Sharma et al., 2021, Oss et al., 2003**). This reduces the use of raw materials and energy, and also the amount of landfilled waste.

The amount of glass waste in Hungary has increased significantly in recent years (**Szép, 2015**). The problem is that most of the generated glass waste is not reused or recycled, as there is no domestic glass factory that accepts it, so the transport costs to foreign glass factories are high. Moreover, enormous quantities of glass waste are disposed, since most of the packaging glass bottles are mixed with other materials (textiles, paper, metal, porcelain, and plastic) in their material structure, and therefore, their treatment is problematic **(Seregély, 2015, Szép, 2015**). In the production of glass foam, this type of glass waste can be used appropriately (**da Costa et al., 2020, Souza et al. 2017, Song et al., 2021**).

With the spread of green technologies, the demand for environmentally friendly insulation materials is growing. Glass foams are widely used in the construction field as structural and thermal-acoustic insulation materials (**Qi et al., 2019, Sharma et al., 2021, Assefi et al., 2021**). Glass foams are porous materials of gas and solid phase, and they have an inorganic chemical structure (**Assefi et al., 2021, Cengizler et al., 2021**). This type of glass recycling has many positive properties, as the glass foam is lightweight, insulating material, frost-resistant, non-flammable, chemically neutral and non-toxic, bacteria-resistant, water and vapour resistant. In addition, glass foam has relatively low transport costs, is easy to handle, and can be easily combined with concrete (**König et al., 2016, Scheffler and Paolo, 2006, Scarini et al., 2005**).

The combination of these properties makes glass foam suitable for a wide range of applications in the construction industry (e.g. for insulating roofs, walls, floors

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Table 1: Chemical composition of glass waste

	SiO_2	Al_2O_3	MgO	CaO	Na_2O	K_2O	Fe_2O_3	MnO	TiO_2	P_2O_5	S	F
Unit	$m/_m\%$	$m/_m\%$	$m/_m\%$	$m/_m\%$	$m/_m\%$	$m/_m\%$	$m/_m\%$	$m/_m\%$	$m/_m\%$	$m/_m\%$	$m/_m\%$	$m/_m\%$
Glass waste	73.6	1.2	2.25	9.89	11.9	0.62	0.47	0.008	0.049	0.014	0.20	<0.3

Table 2: Parameters of the ball mills

Mill type	Amount of grinding media [g]	Amount of raw material (glass waste) [g]	Mill chamber size [cm^3]	Size of the grinding media [mm]	Material of grinding jar and grinding media	Grinding stresses	Vibrating frequency	Rotation rate
Drum mill	7489	660	4710	10 and 20	stainless steel	impact, rubbing, pressure	-	80 rpm
Vibrating mill	1590	141	1000	10	stainless steel	impact	50 Hz	-
Planetary mill	430	43	500	30	stainless steel	impact, friction	-	400 rpm

and ceilings, and as a light weight aggregate in concrete or filler for restoration of failed slopes). Glass foam is produced from glass powder mixed with foaming agent and heat treated at a high temperature (sintering process), where gas formation occurs (**Spence et. al., 2016; Kálnai et al., 2007; Scarinci and Brusatin 2005, König et al., 2016**). During the sintering process, the furnace reaches higher temperatures than the softening temperature of glass where the glass viscosity is inferior to $10^{6.6}$ Pa.s. Foaming agent particles are wrapped by the softened glass until the reaction or decomposition temperature of the foaming agent is reached, it releases gases that form gas bubbles in the softened glass. With the increase of temperature, the glass viscosity decreases, and furthermore the surface tension decreases. The glass decreases the pressure over the gas bubbles and increases the expansion and the pore coalescence as a result of the reduction of surface tension (**da Silva et al., 2021, da Silva et al., 2020, Østergaard et al., 2019**).

In industrial glass foam production, most of the glass is derived from waste glass. The most commonly used glass types are mixed colour container glass bottles and window glass, but there are other types of glass, such as lamp glass and cathode ray tubes (CRT) (**Qi et al., 2019, Saparuddin et al., 2020, Paul et al., 2018, Federico and Chidiac, 2009, Bueno et al., 2020, Attila and Güden, 2013, Li et al., 2013**).

One of the key steps in the production of glass foam is the grinding of the raw materials to the optimum size range. The glass waste is necessary to be ground generally below 100 μm, otherwise the foaming process will not be sufficient. Several studies (**Fernandes et al., 2014, König et al., 2014, Attila et al., 2013**) have shown that there is a relationship between the fineness of the glass powder and the pore diameter of the glass foam.

The aim of this article is to reveal the relationship between the parameters of grinding of glass waste in three various mills with different energy demand to produce glass powder for glass foam in an energy efficient way.

2. Methods

The raw material for the experimental procedure was container glass waste bottles, which were ground in three different ball mills after pre-shredding with a cylindrical mill. During the experiment, the specific grinding energy and specific surface area were determined.

2.1. Methods for raw material preparation and characterization

The raw materials used in the experiment were container glass bottle waste, which contained a significant amount of contaminants, such as paper and organic matter. The first step was to clean the contaminated material in a washing drum and then dry it to mass consistency.

The mixed container glass waste of different colours (white, green, and brown) was pre-shredded using a roll crusher with a screen size of 1 mm. This step was necessary because the particle size of the raw material was too large for the ball mills to be able to grind substantially instead of abrasing the particles.

The particle size distribution of the raw material was measured by dry sieving, based on the measurement x_{10}= 115μm, x_{50}=420μm and x_{80}=800 μm.

The particle size distribution of ground material was measured by HORIBA LA-950V2 laser diffraction particle size analyzer in wet conditions using distilled water as dispersing media applying the evaluation method of Mie theory.

Geometric specific surface area calculation was also obtained by HORIBA LA-950V2 laser diffraction particle size analyzer using Heywood factor 1.

The chemical composition of raw materials was obtained by X-ray fluorescence analysis. **Table 1** shows the results of the chemical analysis of glass waste (XRF).

The chemical composition results (see **Table 1**) of the glass waste showed that these are typical soda-lime glasses, which are used to produce glass bottles.

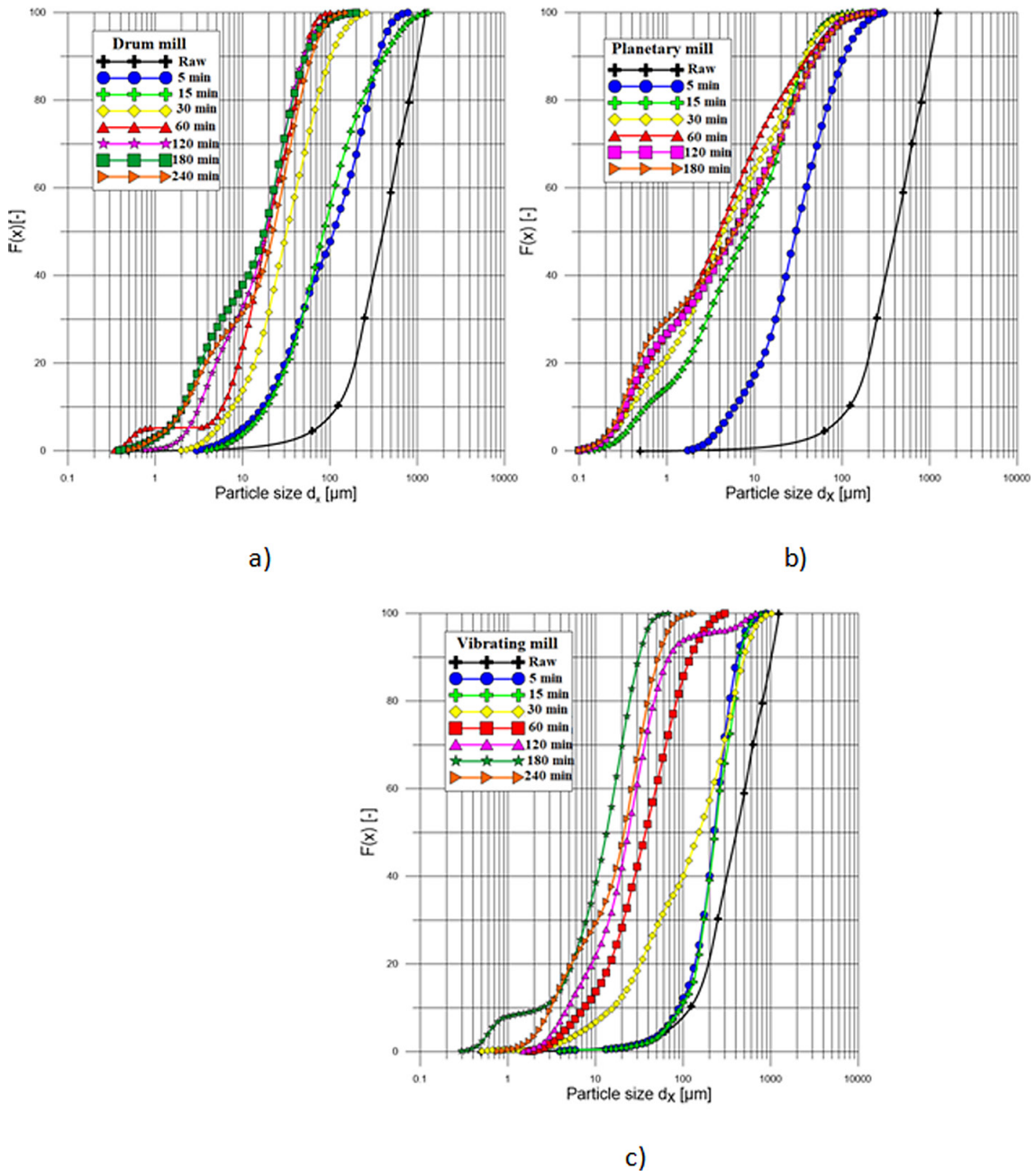

Figure 1: Particle size distribution grinding with the drum mill (a), the planetary mill (b), and the vibrating mill (c)

For characterization of the grindability of glass waste, the Hardgrove Grindability Index (HGI) and calculated Bond - Work Index were used. HGI measurement was obtained by the Universal Hardgrove mill using the Hardgrove test method where the HGI is calculated by **Equations 1 and 2** (**Mucsi 2008, Mucsi, 2019**):

$$HGI = 13+6.93m_{74} \quad (1)$$

where:

HGI – Hardgrove Grindability Index,

m_{74} – mass of the product which below 74 µm (g).

Using the result of HGI the Bond – Work Index can be calculated by **Equation 2** which is the Csőke empirical formula (**Mucsi et al, 2016, Mucsi, 2013**):

$$W_{iB}^{H} = \frac{468}{HGI^{0.82}} \quad (2)$$

2.2. *Methods for systematic grinding tests*

In the systematic grinding experiments, three various mills with different energy intensity are used, namely a drum mill (low intensity), a vibrating mill (medium intensity) and a planetary mill (high intensity).

The filling ratio of the grinding media was 60%, and the filling ratio of the material was 110% of the drum mill and the vibrating mill. One condition of the planetary mill, the quantity of feed material and grinding media were added at a 1:10 ratio, which was determined by the manual of the planetary mill.

Details of the parameters for each mill type are provided in **Table 2**.

3. Results and discussion

Grinding experiments were carried out until the aggregation of the particles in the event of each mill type. The grinding experiments were 5, 15, 30, 60, 120, 180, and 240 minutes of drum mill and vibrating mill. Aggregation occurred in an earlier phase of the milling with the planetary mill, so 240 minutes of grinding was not required. The specific grinding energy was measured using a built-in energy meter for the planetary mill, a single-phase energy meter for the drum mill and a three-phase Carlo Gavazzi digital energy meter for the vibrating mill.

3.1. Particle size distribution

Based on particle size distribution which is provided in **Figure 1 a-c**, the particle size decreased at all events except 240 minutes grinding with the drum mill and the vibrating mill, and 120 minutes with the planetary mill where a significant increase can be observed due to the aggregation. The sample ground for 180 minutes with the drum mill, 180 minutes with the vibrating mill and 60 minutes with the planetary mill had the finest particle sizes with narrow distribution.

Figure 1a) clearly shows that there is no significant difference between 5 and 15 minutes of grinding, but at 30 minutes the particle size decreases significantly compared to the previous two measurements with the drum mill.

Figure 1c) shows that the particle size decreased significantly for the 15 min grinding, but there was no significant change at 30 minutes compared to the previous grindings with the vibrating mill.

Compared to the 5 minutes grinding with the planetary mill (see **Figure 1b**), it can be seen that the particle size has decreased significantly, however, no large differences were observed in the other measurements.

In the grinding kinetics of the drum mill (60 minutes grinding) and the vibrating mill (180 minutes grinding), a part occurred below 1 μm, which disappeared during subsequent grindings.

Table 3 shows the characteristics particle sizes (x_{50}, x_{80}) in the case of each mill type before aggregation occurred.

Table 3 shows that all three mills have characteristics particle sizes (x_{50} and x_{80}) below 100 μm. The smallest particle size was obtained with the planetary mill, followed by the drum mill and then the vibrating mill. Glass grinding aims to grind the glass to the finest possible particle size, in most cases, grinding is supplemented by sieving to achieve the optimum particle size range (**Arriagada et al., 2019, da Costa et al., 2020, Jeong et al., 2019, Lamri et al., 2020, Sapparuddin et al., 2020**). Based on the literature, glass waste is generally ground below 100 μm, for example particle size was below 74 μm used a 74 μm mesh size sieve after grinding (**da Costa et al., 2020**) the average particle size was below 37 μm (**Abdollahi et al., 2020**) and below 100 μm (**Arrigada et al., 2019, Jeong et al., 2019, Lamri et al., 2020, Liu et al., 2019, Polat and Güden, 2021, da Silva, 2019, Smiljanić et al., 2021**).

Table 3: Characteristics particle sizes of each mill type before aggregation

Mill type	Grinding time [min] (before aggregation)	X_{50} [μm]	X_{80} [μm]
Drum mill	180 min	13.39	15.41
Vibrating mill	180 min	17.61	23.49
Planetary mill	60 min	3.99	13.83

3.2. Specific surface area and characteristic particle sizes

The particle size distribution and the specific surface area were determined using a Horiba LA-950V2 laser diffraction particle size analyzer. **Figure 2a-c** shows the specific surface area (SSA) and x_{50}(median particle size), x_{80}(80% of particle size) values as a function of grinding time for each mill type.

The specific surface area values of all the samples obtained from the three mills rose with an increase in the grinding time. Wide-ranging specific surface areas were obtained of grinding with the planetary mill, followed by the drum mill, which had much lower values, and then the vibrating mill. Furthermore, it can be seen that as the grinding time increases, the values of x_{50} and x_{80} particle size decrease, and the specific surface area increases, respectively. The longer the grinding time, the smaller the particle size and the larger the specific surface area is observed until aggregation, where the particle size increases. There is no significant change in the particle size x_{50} and x_{80} from 60 minutes grinding until 240 minutes grinding of drum mill, but the specific surface area increases due to the grinding stresses during milling affecting mainly the smaller particles.

3.3. Specific grinding work

The specific grinding work of the three ball mills was measured with energy meters. The specific grinding work can be calculated using **Equation 3**.

$$W_{spec.} = (W_{obtained} - W_{no\text{-}load})/m_{material} \quad (3)$$

Where:

W_{spec} – Specific grinding work,
$W_{obtained}$ – Obtained energy,
$W_{no\text{-}load}$ – No-load energy,
$m_{material}$ – Amount of the feed material.

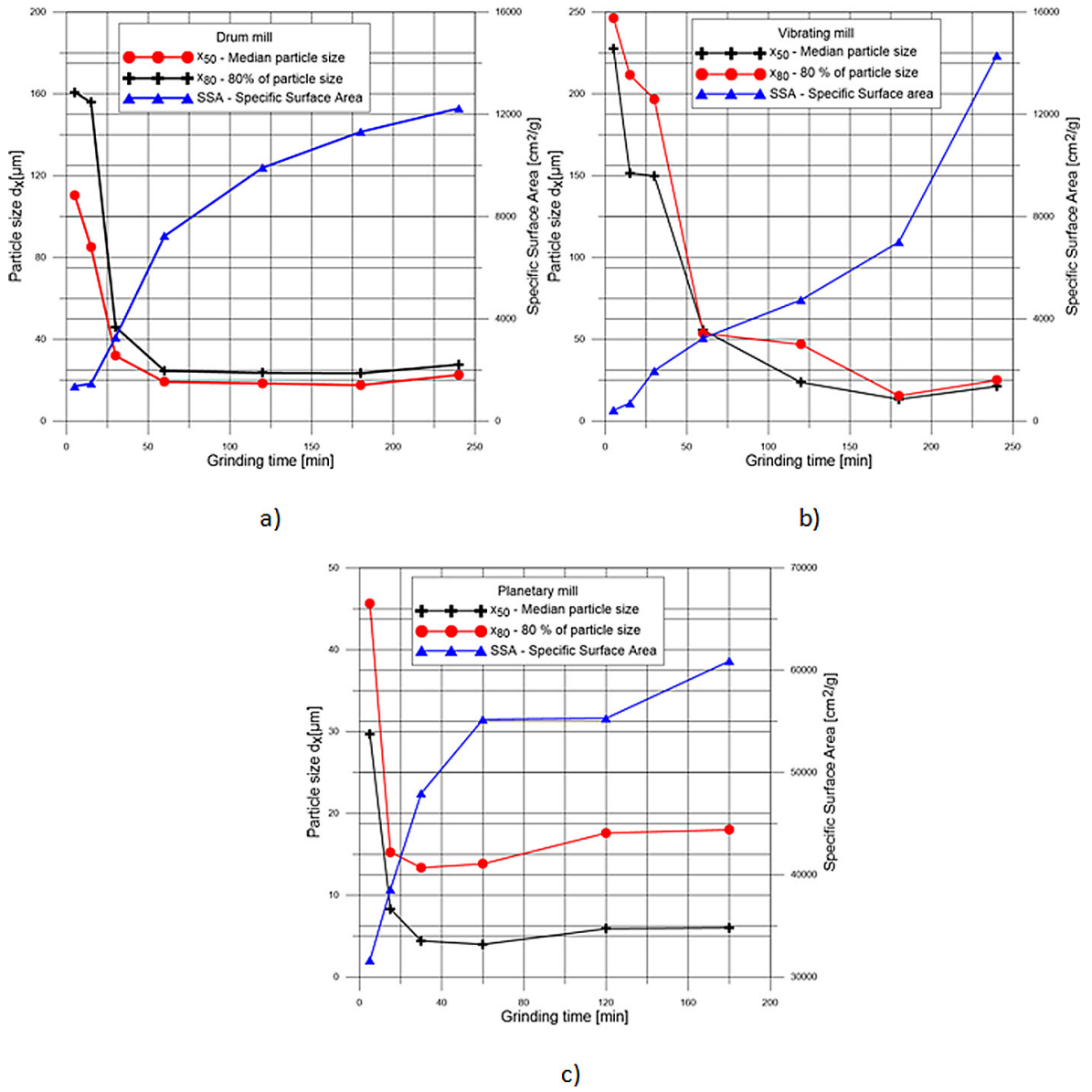

Figure 2: Characteristics particle sizes (x_{50} and x_{80}) and specific surface area (SSA) values in case of grinding with the drum mill (a), the vibrating mill (b), and the planetary mill (c)

Figure 3 shows the specific grinding work of the mills as a function of time. It can be seen that the planetary mill has the highest specific grinding energy, followed by the vibrating mill and then the drum mill. There is not much difference between the vibrating mill and the drum mill, but the planetary mill requires much more energy compared to the other two types of mills.

3.4. Hardgrove Grindability-Index (HGI) and Bond – Work Index

The Hardgrove Grindability Index was carried out from five parallel measurements. The Bond – Work Index calculated from the obtained HGI value using the Csőke empirical formula (**Equation 2**) (**Mucsi, 2013, Faitli et al., 2017**) which can be seen in **Table 4:**

The calculated Bond – Work Index should be corrected because of the deviations from the model conditions with the following formula (**Mucsi, 2009**):

$$W_{iBcorr} = kW_{iB}^{calculated} \quad (4)$$

The k factor is used to take into account circumstances other than the Bond procedure, where $k=k_1+k_2+k_3+k_4+k_5+k_6$, are the correction factors developed by Bond.

The relevant correction factors are:

- k_1: dry grinding factor, the value is 1.3
- k_5: fine grinding factor, if 80 % of particle size is < 75 µm $k_5 = \frac{x_{80}+10.3}{1.145 * x_{80}}$
- k_6: low degree of comminution, when r is <6, then $k_6 = 1+\frac{0.13}{X_{80} / x_{80-1.35}}$

Table 4: HGI and calculated Bond – Work Index values

Test	Hardgrove Grindability Index [-]	HGI derived Bond – Work Index [kWh/t]
1.	48.343	19.457
2.	48.135	19.526
3.	47.512	19.737
4.	47.442	19.761
5.	47.997	19.573
Average	**47.886**	**19.611**

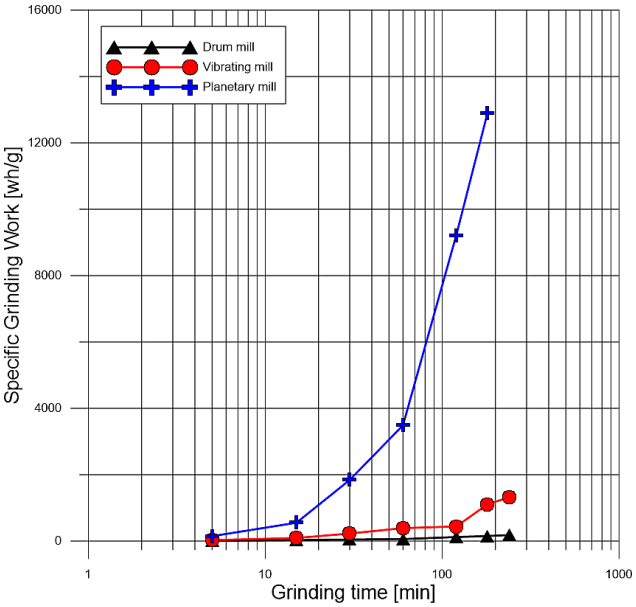

Figure 3: Specific grinding work of each mill type

The Bond - Work Index obtained by taking into account the correction factors was 32.302 kWh/t. Against the correction factors, k_1, k_5, and k_6 were necessary to multiply the previously calculated Bond Work Index from the HGI number, the other factors were not relevant for the measurement. The value of the k_1 factor was given, which was 1.3, and the k_6 factor was calculated by determining the degree of comminution and using the given formula, whose result was 1.267. The corrected Bond Work Index was multiplied by the fine grinding factor (k_5) of each mill type.

Table 5: Fitted power function equations and correlation coefficient values of corrected Bond – Work Index values

Mill type	Equation of fitted power function	R^2
Drum mill	y=-0.115 * ln(x) + 4.044	0.909
Vibration mill	y=-0.111* ln(x) + 4.042	0.788
Planetary mill	y=-0.293 * ln(x) + 4.655	0.991

Figure 4 shows the Corrected Bond – Work Index values from HGI result. It can be seen that for each mill type, the longer the grinding time and the smaller the particle size, the higher the value of the corrected Bond

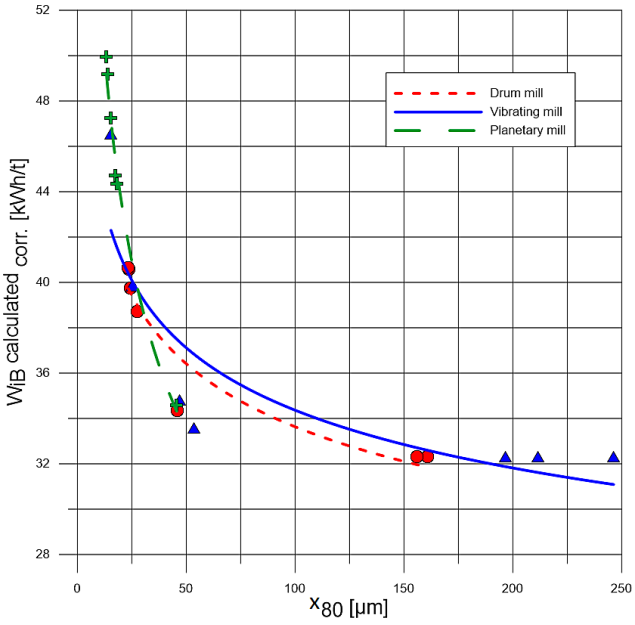

Figure 4: Corrected Bond – Work Index values from HGI

– Work Index. It can be concluded that after aggregation, this value started to decrease again.

Table 5 shows the equations and correlation coefficient (R^2) values of the power functions fitted to the corrected Bond – Work Index values. The lowest correlation coefficient (R^2) value occurred of the vibrating mill, followed by the drum mill, and the planetary mill however it can be seen that the correlation coefficient (R^2) values of the drum mill and the planetary mill are fitted well compared with the vibrating mill's value.

4. Conclusions

The following conclusions were drawn from the obtained results:

(1) The planetary mill has the highest specific grinding work, followed by the vibrating mill and then the drum mill for a given material fineness.

(2) Corrected Bond – Work Index of the product and characteristic particle size (x_{80}) relationship can be described by a power function. The correlation factor was relatively high in the case of the drum mill and the planetary mill.

(3) The obtained specific surface area (SSA) values show an upward trend with an increase in the grinding time. The planetary mill achieves the highest specific surface area, followed by the drum mill and the vibrating mill.

(4) The characteristic particle sizes (x_{50} and x_{80}) show a downward trend with an increase in the grinding time of each mill type until aggregation, where a significant increase can be observed.

(5) Aggregation occurred at different grinding times, i.e. 60 minutes of grinding with the planetary mill, 180

minutes of grinding with the drum mill and the vibrating mill.

(6) As a result, 180 minutes of grinding with the drum mill is chosen as the optimal setting for glass grinding in the given size range in consideration of the economic benefits, like low energy demand and low specific grinding work.

(7) Furthermore, the resulted specific surface area is relatively high and if industrial quantities of glass waste are necessary to grind, the drum mill has a much higher capacity compared to the vibrating mill and the planetary mill.

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SAŽETAK

Usporedno istraživanje mljevenja staklenoga otpada primjenom različitih vrsta mlinova

Ovaj rad bavi se sustavnim ispitivanjem mljevenja i određivanjem mogućnosti mljevenja staklenih boca. Sustavna ispitivanja mljevenja provedena su u trima različitim vrstama kugličnih mlinova (bubnjasti mlin, vibracijski mlin i planetarni mlin) različitih energetskih zahtjeva (niskoga, srednjega i visokoga intenziteta) u suhim uvjetima. Osim toga, kod svakoga ispitivanja određena je specifična energija mljevenja i specifična površina. Ispitivanje meljivosti provedeno je pomoću mlina Universal Hardgrove, a Bondov radni indeks izračunan je iz indeksa meljivosti po Hardgroveovoj metodi. U ovome istraživačkom radu fokus je bio na analizi meljivosti staklenih boca, a cilj je bio energetski učinkovito mljevenje staklenoga otpada kako bi se proizveo prah staklene pjene za daljnju uporabu (npr. staklena pjena) na održiv način. Na temelju dobivenih rezultata može se zaključiti da je optimalni mlin za mljevenje staklenih posuda bubnjasti mlin zbog toga što ima nisku specifičnu energiju mljevenja (151,52 kWh/t) u usporedbi s drugim dvjema vrstama mlinova (3488,37 kWh/t planetarnoga mlina i 1106,38 kWh/t vibrirajućega mlina), što rezultira relativno visokom specifičnom površinom (11 314 cm^2/g). U slučaju mljevenja industrijskih količina staklenoga otpada bubnjasti mlin ima znatno veći kapacitet u odnosu na vibracijski i planetarni mlin.

Ključne riječi:
stakleni otpad, reciklaža, mljevenje, kuglični mlin, specifična energija mljevenja

Author's contribution

Ildikó Fóris (1) (PhD student) performed the laboratory work, writing and editing. **Gábor Mucsi (2)** (Professor) conceptualization, formal analysis, and editing.