

Received December 3, 2018; reviewed; accepted January 14, 2019

Adjustment of limestone grinding in an electromagnetic mill for use in production of sorbents for flue gas desulphurization

Marta Wołosiewicz-Głąb ¹, Paulina Pięta ², Dariusz Foszcz ², Tomasz Niedoba ², Tomasz Gawenda ²

¹ The Mineral and Energy Economy Research Institute of the Polish Academy of Sciences, Krakow, Poland

² AGH University of Science and Technology, Faculty of Mining and Geoengineering, Department of Environmental Engineering and Mineral Processing, Mickiewicza 30 Av., 30-059 Krakow, Poland

Corresponding author: wolosiewicz@gmail.com (Marta Wołosiewicz-Głąb)

Abstract: The paper presents the study of the effectiveness of the grinding in an electromagnetic mill for limestone with the feed particle size up to 0.5 mm and 1 mm. The goal was to prepare material for specific particle size fractions of fine and coarse sorbents used for flue gas desulfurization. The work focused on optimizing the duration of the grinding and selecting grinding media properties to obtain the highest relative increase in the 50 µm and 50-100 µm particle size fraction in the grinding product. An important element for grinding control is the knowledge of the impact characteristics for main parameters and factors on the efficiency of the material comminution. The grinding results show its kinetics of grinding product yield. A model was also created that shows the relationship between grinding time and the process efficiency and can be used to optimize the process. The research allowed to determine the impact of changes in the parameters of the mill and the feed, which will allow to determine the controls for the system's continuous operation. It is crucial to determine the efficiency of the mill. It depends on the degree of material fragmentation and the granularity of the feed subjected to the grinding. Both of these parameters are different for different raw materials. Determination of grinding kinetics models allowed to determine the dependence of function between the growth of the selected particle size fraction in the product and the residence time of material.

Keywords: mineral processing, electromagnetic mill, limestone grinding, sorbent for flue gas desulphurization, process modelling, grinding kinetics

1. Introduction

Comminution of the raw material is an operation widely used in many industries from mineral processing through the chemical, constructions, food, cosmetic and pharmaceutical industries. It is an energy intensive process which efficiency improvement might change the operating costs of the mill. That is one of the reasons for many scientific researches developed in order to increase comminution efficiency. Starting with Rittinger, Kick, Bond and Walker and Shaw (Rittinger, 1987; Kick, 1885; Bond, 1952; Walker and Shaw, 1954), where energy-particle size relationship was described, there are studies that focused on the optimization of grinding effectiveness and energy consumption for conventional mills. Among others, an interesting example is the work of Delboni and Morrell, whose research developed a new autogenous and semi autogenous (AG/SAG) mill model that is based on charge dynamics. The model relates charge motion and composition to power draw and size reduction. Size reduction is described by considering impact and attrition/abrasion as separate processes. These are linked to the energy available in the mill, the charge size distribution, and the relative motion of the grinding media (Delboni and Morrell, 2002).

Also work of Shin et al. (2013) investigates the effect of ball size and powder loading on the milling efficiency. Their research is based on wet-milled alumina powder by zirconia balls with varying rotation

speed. They proved that as the rotation speed increases, kinetic energy of the ball increases, which results in shifting the optimum ball size toward a smaller value. It is interpreted as the result of competition between the reduced kinetic energy of the colliding balls in the slurry (a negative source of milling efficiency) and the increased number of contact points of the smaller balls (a positive source for milling efficiency), which yields the optimum ball diameter at an intermediate ball size (Shin et al., 2013).

Another research on modelling improvement of the grinding done by Petrakis and Komnitsas (2017) is based on the use of matrix and population balance models. Such interesting approach identifies natural events during grinding. Combination of models (where each of them has its limitations and capabilities) offers some advantages. Matrix model and the selection function were combined to predict the size distribution of the ground products (Petrakis and Konitsas, 2017)

As shown, many scientific researches on grinding efficiency are based on different modelling methods. In mineral processing the main goals for modelling are usually connected either with process optimization issue or mechanism study. These tasks are not easy because the processes are usually not stable and they depend on many features which monitoring is limited and not entirely precise. Although the necessity of searching for such models is crucial.

The simplest way of searching for the solution and/or equation describing the process is regressive methods. The use of various statistical techniques is conditioned by the goal assumed during analysis and modelling as well as empirical data characteristics. In case of regression analysis, they must meet certain conditions. It is necessary to check the following assumptions: dependence is linear; no significant outliers; homoscedasticity – the variance of reminders of the random component is equal for all observations; the reminders have a distribution similar to the normal distribution. For multiple regression, following conditions have to be satisfied: the number of observations must be greater than or equal to the number of parameters, the parameters are not collinear, there are no auto correlated residues. The researcher can also apply the data actualizations, adjustment techniques like Moving Average (MA) or Exponentially Weighted Moving Average (EWMA), dependably on sort of data, number of parameters and quantity of the data. In more complicated cases other methods can be arranged like data mining techniques, neural networks, genetic algorithms, non-linear programming etc. (Berthouex and Brown, 2002).

However, in industrial practice the simplest solutions are usually the best. In this paper, the various exponential regressive modelling approaches are proposed to determine increments in 0-50 and 50-100 μm particle size fraction for the limestone comminution products in an innovative electromagnetic mill (EMM). Furthermore, the basic information regarding energy consumption is presented in relation to grinding time. The analyses were performed for various grinding media combinations.

Limestone sands and powders are widely used in the production processes of building materials, water purification, flue gas desulphurisation and other areas of the economy. The purpose of comminution is to obtain products with a specific particle size, shape and surface area (especially in cement industry (Touil et al., 2008)).

Sorbents are porous materials used to collect, retain and absorb, above all, various types of liquids such as: oils, petroleum substances, acids, alkalis, water and other liquid substances. Sorbents can be all substances that have the ability to retain toxic liquids inside (absorption process) or on their surface (adsorption process), and also use both phenomena simultaneously (Chodorowski and Salomowicz, 2004). The article focuses on preparing the feed for limestone sorbents (limestone powders and sands, in some cases also burned lime or hasraticated lime) constitute the largest group of reagents used in flue gas desulphurisation installations in Poland. Flue gas desulphurization is a set of technologies used to remove sulphur dioxide (SO_2) from exhaust flue gases of fossil-fuel power plants, and from the emissions of other sulphur oxide emitting processes (e.g. trash incineration).

At the moment high-quality limestone, exploited from deposits of various ages, is used for the production of lime sorbents used to reduce SO_2 emissions in Poland. The research was aimed at assessing the efficiency of the mill for various grinding media types and feed particle size in order to determine its operating parameters. It supplemented the existing research described in Ogonowski et al., 2018; Ogonowski et al., 2017; Pawełczyk et. al., 2015 and Wołosiewicz-Głąb et al., 2018 in the field of grinding efficiency evaluation in an electromagnetic mill. The research allowed to determine the

parameters of the mill's operation to achieve optimal effects of material fragmentation. The novelty presented in the article is based on the definition of dependence between the parameters of the grinding in the electromagnetic mill for various grinding media and feed particle size distribution. It enables to determine grinding efficiency depending on feed parameters and choosing the right set of grinding media for the expected process results. The paper focuses on the analysis and modelling of yields (1) and increments of particle size fractions (2) 0-50 and 50-100 μm , which are the key material for the production of limestone sorbents for the purification of exhaust gases.

Process modelling allows to design and execute algorithms used in the electromagnetic mill control system. They enable to control on going analysis of particle size distribution and the operating status of the mill and recycle. With a layer of algorithms, properly designed measurement equipment defines the parameters of the final product quality and energy consumption. Models would also allow the next step in development program based on the simulation studies and industrial impact of physical parameters scaling on the individual parameters of the quality and efficiency.

2. Materials and methods

Limestone powder below 120 μm , which are used in the wet lime method are mainly used for desulphurisation during combustion in fluidized bed boilers (Chodorowski and Salomowicz, 2005; Kotowski and Ratajczak, 2010; Hlincik and Buryan, 2013). The following tests were carried out on two limestone feed types with particle size 0–0.5 mm (fine) and 0-1 mm (coarse). The material originated from the "Czatkowice" Limestone Mine, Czatkowice, Poland. Limestone was characterized by a stable content of CaCO_3 . The chemical composition of the tested samples was determined after dissolution using a microwave mineraliser in a mixture of hydrochloric acid, nitric and hydrofluoric acid. The obtained solutions were analysed with ICP-OES spectrometer to determine the level of concentration of the elements. Results of the analysis are presented in Table 1. The material whiteness was also constant (a physical property that determined the use of minerals, inter alia as a sorbent). This feature results, in part, from the fact that the tested limestone contains a small number of components that may affect the change or decrease of this parameter value. One of them is Fe_2O_3 . Also, the degree of dolomitization in the test sample is not large with amounts of MgO 1.25% with the limit values up to 1.78% (Kotowski and Ratajczak, 2010).

In chemical composition of the sample, 50,08 PPM of As was found. It might raise the question about possible environmental concerns when limestone is used as a sorbent and discarded after the end of life. In order to neutralize this type of waste, processes of precipitation of sparingly soluble heavy chemicals compounds contained in waste to reduce their solubility in water and processes of their immobilization by closing in a concrete mass, sintering in ceramic materials or vitrification are being used.

The mill type, operating parameters such as media size, shape, mill speed, and powder loading (Gupta and Sharma, 2014) define the degree of fragmentation to be achieved. Grinding is also used to pulverized mineral aggregates, and the efficiency is strictly dependent on the type of feed.

Production of fine granular materials intended for sorbents for flue gas desulphurization is carried out in various mills and classifiers constituting technological systems of mechanical processing. Mechanical and chemical properties of the raw material (feed – limestone) as well as the required particle size below 120 μm defines the selection of the technological system along with the type of mill, which makes the electromagnetic mill technology particularly useful in the case of fine grinding with P_{80} below 30 μm or when obtaining such a fine size fraction would require disproportionate energy expenditure.

Finer the desired product, the more energy should be put into its comminution, the optimal fineness can be obtained by increasing the grinding time, changing grinding media, reducing the feed particle size (by crushing or pre-grinding in the earlier stage of processing) or by introducing additional grinding or changing the type of mill (Napien-Munn and Wills, 2006; Wand and Forssberg, 2007). Table 2 presents the specific energy consumption in relation to the grinding time in the electromagnetic mill. This table shows that energy changes with the longer duration of the process. The energy consumption results from the mass of ground material and energy consumption by the mill. Data presented in Table

2 are brief due to the scope of this work. More information can be found in Ogonowski et al. (2018) and Krawczykowski et al. (2018).

Table 1. Chemical composition of the analysed limestone feed

Element	Content
SiO ₂	1,77%
TiO ₂	0,03%
Al ₂ O ₃	0,56%
Fe ₂ O ₃	0,35%
CaO	52,39%
MgO	1,25%
MnO	0,02%
K	0,09%
K ₂ O	0,10%
Na	0,02%
Na ₂ O	0,02%
SrO	0,03%
S	0,03%
P	0,05%
H ₂ O	0,07%
C _{org}	0,84%
CaCO ₃	93,50%
C _{inorg}	11,22%
MgCO ₃	2,62%
Metals	Parts per million [PPM]
As	50,07
Cd	2,56
Cr	7,53
Cu	22,80
Hg	2,96
Ni	5,40
Pb	77,37
Zn	188,43

Table 2. Specific energy consumption in comparison to grinding time in D200 electromagnetic mill caption

Grinding Time (s)	Energy Consumption (kWh)	Specific Energy (kWh/t)
5	0.025	50
10	0.05	100
15	0.075	150
20	0.1	200
25	0.125	250
30	0.15	300

In order to determine the equations describing the output dependence function of the considered particle size fraction on the grinding time, a regression analysis was carried out. Also, the accuracy of matching the generated equations to the empirical data was checked. The considerations concerned the linear and quadratic functions, the polynomials of higher degrees and the logarithmic function. In the paper, the best results of modelling were presented according to the match ratios. The remaining cases were rejected due to the unsatisfactory level of real data mapping. Analysis included two different approaches but the fit was similar and increment curves have similar shape.

In order to quantify the relationship between grinding time and the mentioned indicators, approximation using the method of least squares was carried out, the exact description of which can be found in the other papers (Foszcz, 2006; Berthouex and Brown, 2002; Lee-Ing and Chung-Ho, 2002).

$$\gamma_i = \frac{m_i}{\sum_{i=1}^n m_i} \quad (1)$$

$$\Delta(d_i) = \frac{a_{pi} - a_{ni}}{100 - a_{ni}} \quad (2)$$

where: a_{pi} – a mass of the i -th particle size fraction in the milling product,

a_{ni} – a mass of the i -th fraction in the feed.,

m_i – a mass of a product.

$\Delta(d_i)$ – increments of particle size fractions 0-50 and 50-100 μm

γ_i – a yield of a product

The functions of dependence of the yield of the studied particle size fraction y_i on grinding time were determined by means of polynomials of the second and third degree, the form of which is given by the general formula (3):

$$y(x) = a_0 + a_1 \cdot t^n + a_2 \cdot t^{n-1} + a_3 \cdot t^{n-2} + \dots + a_n \cdot t \quad (3)$$

where: a_i – model coefficient,

t – grinding time.

The approximation of the presented increment curves of the particle size fractions for the grinding in an electromagnetic mill was based on the distribution of a single parameter exponential distribution (4):

$$\Phi(t) = \begin{cases} 100 \cdot (1 - e^{-\lambda t}) & \text{for } x > 0 \\ 0 & \text{for } x \leq 0 \end{cases} \quad (4)$$

where: λ – coefficient of the exponential function,

t – grinding time.

Fundamentally, an exponential distribution is used to model phenomena and changes occurring at a given time. In this work, it was used to illustrate the increase in the content of 0-50 and 50-100 μm particle size fractions in final products obtained by comminution of the limestone in an electromagnetic mill (Tumidajski, 1997; Tumidajski and Saramak, 2009; Niedoba, 2013).

In order to assess the accuracy of matching the regression functions to the experimental data, a mean squared error and a coefficient of determination were used. They are described by formula (5) and (6) (Cameron and Hangos, 2001; Maxwell and Delaney, 2004; King, 202)

$$MSE = \sqrt{\frac{1}{n-k} \sum (y_{ei} - y_{ti})^2} \quad (5)$$

$$R^2 = \frac{\sum_{i=1}^n (y_{ei} - \bar{y})}{\sum_{i=1}^n (y_{ti} - \bar{y})} \quad (6)$$

where: y_{ei} – an empirical value of parameter,

y_{ti} – a theoretical value of parameter,

\bar{y} – an arithmetic mean of empirical values of Y – random variable,

n – number of cases,

k – number of variables in the model.

Grinding in electromagnetic mill is different than in conventional mills. The material is being fed from the top to the vertically positioned working chamber of the mill, directly above the working space of the mill filled with rotating grinding media. The main stream of the gaseous medium in which the grinding takes place and which forces the final product to be received is fed from the bottom of the

mill's working chamber, below the working space of the mill filled with rotating grinding media, preferably under pressure, ensuring vertical movement in the mill's working space (Ogonowski et al., 2018; Ogonowski et al., 2017).

In the case of a mill in a recirculating system, the stream of material recycled with an additional stream of gaseous medium is introduced from the bottom of the working chamber of the mill, below the working space of the mill filled with rotating grinding media. The size of streams of recycled material and additional gaseous medium is controlled independently to feed stream and the main stream of the gaseous medium (Pawelczyk et al., 2015; Wołosiewicz-Głąb et al., 2018). In the case of a coarse material in the feed, such as impurities or oversized grains, it is picked up from the bottom of the working chamber of the mill, i.e. below the working space of the mill. Fig. 1 presents the grinding and classification circuit of the electromagnetic mill.

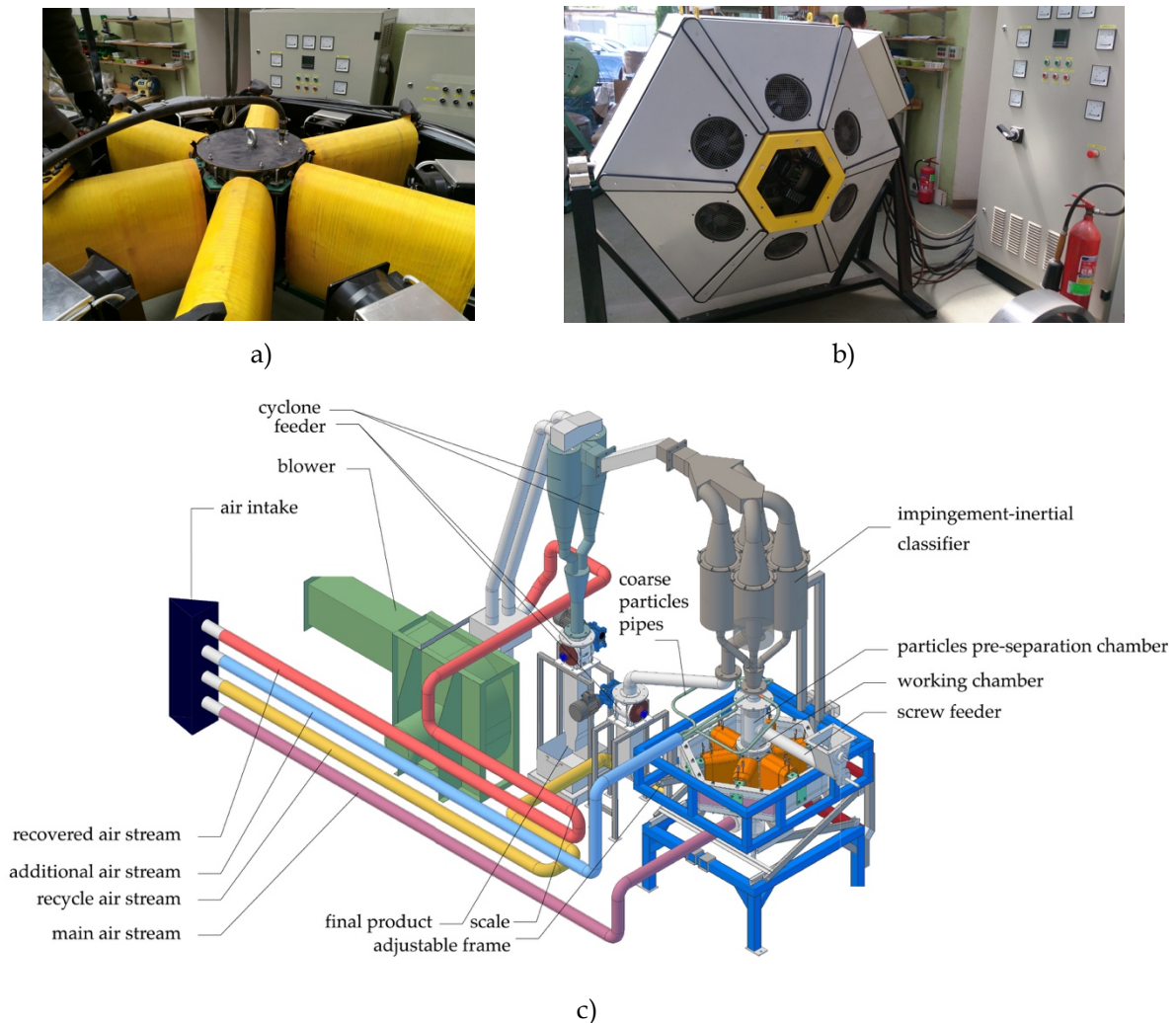


Fig. 1. Electromagnetic mill a) without cover, b) EMM and the control board, c) grinding and classification simplified diagram of the electromagnetic mill. Scheme courtesy of ELTRAF Co., Lubliniec, Poland (Ogonowski et al., 2018)

Physical parameters of the grinding media depend on the mill diameter and the feed particle size distribution. The most common size is 1–3 mm diameter and 10–15 mm length. The shape and size for particular application are being developed by simulations using finite element analysis in order to get the optimum effectiveness of electromagnetic field usage. The grinding media are made of steel (1H18N9) and were provided by Goodsteel, Nowe Ząbubice, Poland.

A laboratory set-up for batch grinding was designed by ELTRAF Co., Lubliniec, Poland. A batch working chamber for electromagnetic mill D200 in the form of a non-ferromagnetic capsule was built.

Its cylindrical shape with one removable base allows inserting the material and grinding the media batch. The base cover is firmly closed, and the capsule is inserted into the mill inside the electromagnetic (EM) field inductor. The second base of the capsule has a small opening that is connected to the pressure release valve for safety reasons. The mill itself is equipped with a supply and supervisory cabinet that allows the monitoring and control of process parameters. A laboratory is also equipped with a scale and moisture analyser that allows precise preparation of dry batches (Ogonowski et al., 2018; Wołosiewicz-Głąb et al., 2018). For the experimental purpose material batches of a limited size range were prepared.

A precise batch of grinding media can be prepared with certainty that they will not leave the working chamber during experiments.

In the study, 24 limestone samples with a weight of 500 grams each were dry milled. The material volume was 4% of the working chamber volume (WCV). Grinding was carried out in six processing times: 5, 10, 15, 20, 25 and 30 seconds. Four types of grinding media were tested (length (mm)/diameter (mm)): 12/2, 12/1.5, 10/1 and additionally a mixture of different sizes (0.75 kg of 12/1.5 media; 0.45 kg of 10/1 media and 0.3 kg of 12/2 media) was also used. During each experiment, the grinding media weight was 1500 grams, which was 9% of the WCV. All samples were subjected to a dry mechanical granulometric analysis. The sieve analysis was carried out on sieves measuring 1, 0.63, 0.5, 0.2, 0.125, 0.071 mm and the measurements were repeated four times to maintain the required representativeness of the results.

Table 3 presents a comparison of an average results for 0-50 μm class yield in grinding products. Repeatability of results obtained under the same conditions determined by the mean value of standard deviation ranged between 3.93-15.10% in relation to the increments of individual particle size fractions. Each measurement was repeated five times.

Table 3. Yield of 0-50 μm particle size fraction in limestone grinding products with fine and coarse feed

Feed size [mm]	Grinding media type (designation)	Yield of 0-50 μm particle size fraction at the specified grinding time					
		Time [s]					
		5	10	15	20	25	30
Fine: 0-0.5	12/2 (1)	67.1	84.9	93.4	94.9	96.7	96.4
	12/1.5 (2)	77.8	87.2	93.8	96.9	97.6	99.5
	Mix (3)	79.2	89.5	96.6	98.6	99.5	99.6
	10/1 (4)	86.2	94.5	98.0	99.0	99.8	99.8
Coarse: 0-1	12/2 (5)	67.9	83.7	89.6	92.4	94.4	97.0
	12/1.5 (6)	70.7	85.3	88.9	95.6	97.8	97.6
	Mix (7)	73.2	89.6	93.3	97.8	98.9	98.8
	10/1 (8)	75.7	93.8	97.7	99.5	99.5	99.6

It can be seen that the grinding results obtained with both feeds are similar. The key factors in controlling the material grinding in an electromagnetic mill are: grinding time and media. The shape of the increment curves confirms that in the majority of analysed cases the highest yield for fine feed can be observed in the first 15 seconds of grinding using 10/1 mm grinding media.

3. Results and discussion

The results of the experiments make it possible to determine models describing the growth of 0-50 and 50-100 μm particle size fractions and their yields in the products of limestone grinding in electromagnetic mill. Fig. 2 shows the models of the outflow of the aforementioned particle size fractions for the investigated cases. They were described using the second-degree polynomials using the least

squares method. The results obtained for the two types of feed tested during the experiment were similar and there was no significant effect of this factor on the final results.

The assessment of the accuracy of model fitting to empirical data was based on the MSE and R^2 index. The results are summarized in Table 4. Based on them, it can be concluded that in general terms, models are characterized by a high degree of convergence with empirical data. This is also confirmed by the results of the standard deviation s_i , where $i \in \{5, 10, 15, 20, 25, 30\}$, presented in the diagrams (Fig. 2).

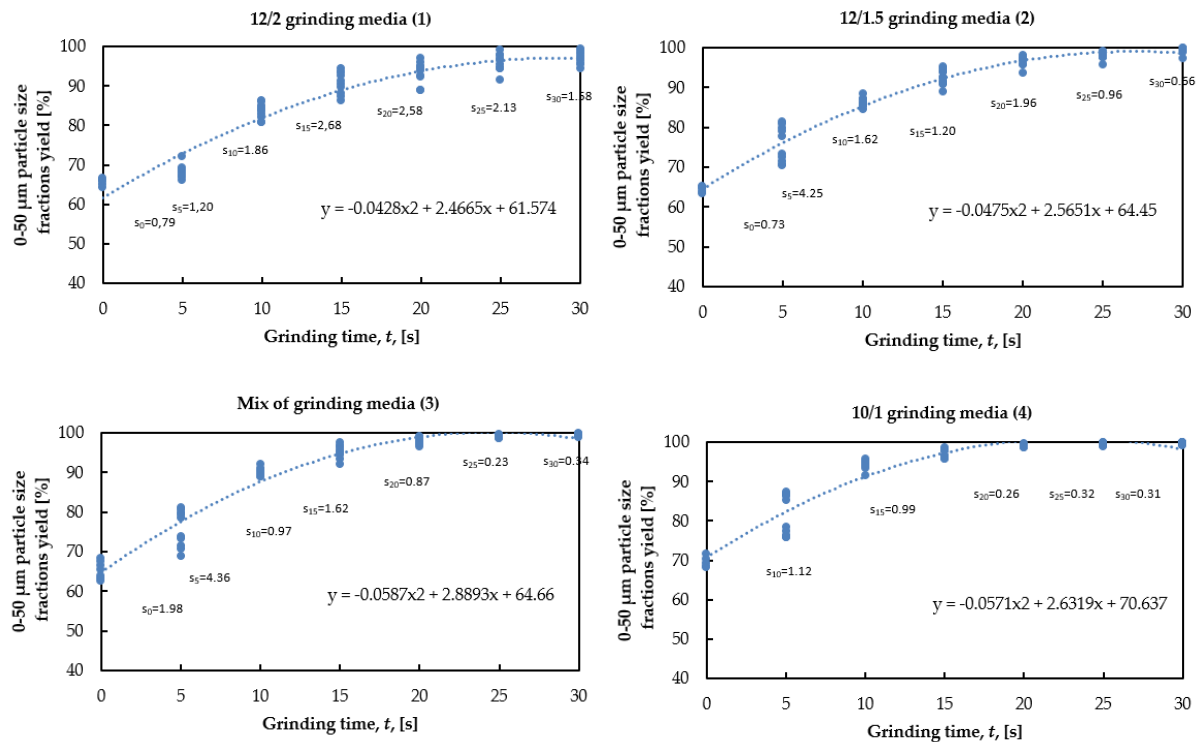


Fig. 2. Models of the 0-50 μm particle size fractions yield (axis y) determined for particular grinding media types (1), (2), (3), (4) depending on time (axis x) taking into account fine and coarse feed average mean values

Table 4. Evaluation of the model fitting for 0-50 μm size fraction for average mean values fine and coarse feed

Grinding media type	MSE [%]
12/2	2.66
12/1.5	2.15
mix	2.40
10/1	2.47

Considering the grinding energy consumption, it is worth to investigate the time dependence on the relative increase of the studied particle size fractions. For this purpose, calculations were carried out in accordance to the Equation 3 (4-increment). Models based on exponential functions were prepared. The results are presented in Fig. 3 in relation to the fine feed and in Fig. 4 for coarse feed.

Regardless of the feed particle size, it can be stated that the largest increment of the 0-50 μm particle size fraction was noticed when using 10/1 grinding media. For limestone comminution, the largest increase in the desired particle size fractions is recorded in the first 10 seconds. From 15 sec., the relative increase in the 0-50 μm particle size fraction is not so significant. The models presented in Fig. 3 and Fig. 4 are described by general formula of the distribution of a single parameter exponential distribution (Eq. 4) and the λ coefficients are presented in Table 5. The standard estimation error in the aforementioned cases is satisfactory – in average 4.22% for coarse feed and 3.96% for fine feed (Table 5). The models give an overview on the kinematics of the grinding described by the yield and the

increments of 0-50 and 50-100 μm particle size fractions. They allow to assess at what stage the comminution should be interrupted in order to obtain the best results. It translates into the maximization of the yield of interesting size fractions, taking into account energy consumption, which is closely related to the optimization of the process time.

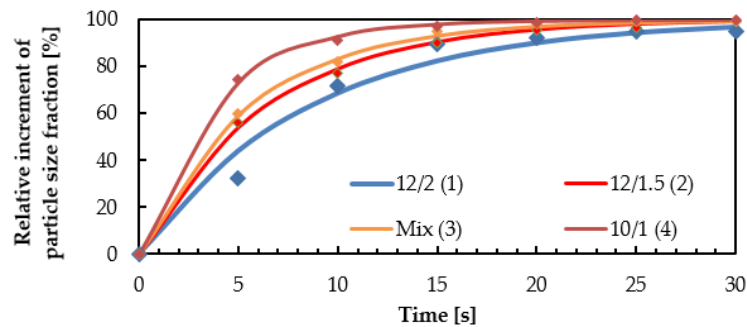


Fig. 3. Models of relative increase of 0-50 μm particle size fraction in grinding of fine feed with grinding media sets (1), (2), (3), (4)

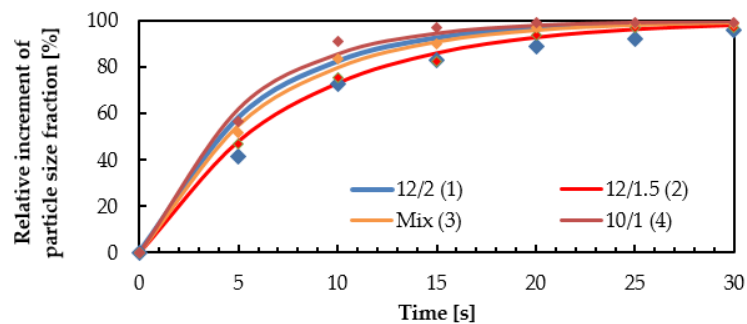


Fig. 4. Models of relative increase of 0-50 μm particle size fraction in grinding of coarse feed with grinding media sets (1), (2), (3), (4)

Table 5. Evaluation of model fitting based on the value of the standard estimation error

Grinding media type	Standard estimation error [%]	
	Particle size fraction	
	0-1	0-0.5
12/2	3.85	10.17
12/1.5	3.44	2.19
mix	3.68	1.81
10/1	5.91	1.66

By analysing the λ values for the generated increment models, one can state certain dependencies, which are shown in Fig. 5.

The increase in the value of the λ coefficient is the same as the increase in the particle size fraction $\Delta(d_{0-50})$ for the grinding time t . For a coarse feed the points arrange in a characteristic way, showing a linear relation between the ratio λ and $\Delta(d_{0-50})$ with a high degree of R^2 fit – over 99%. In the case of fine feed, a more random nature of the relationship between λ and $\Delta(d_{0-50})$ is noticeable. Points are arranged irregularly and curvilinear. Model fit to empirical data is lower and amounts to 86%. In this case, the model has been described by the quadratic equation presented in Fig. 5. Previous attempts to generate an equation based on a linear, power, logarithmic or exponential function gave much worse results. However, in both cases, it can be concluded that with increase of the λ value the particle size fraction 0-50 μm yield increases as well.

Under the same conditions, limestone comminution tests were also carried out with the aim of obtaining the 50-100 μm size fraction. The yield of this particle size fraction was much smaller than in the case of the 0-50 μm class. It is due to the particle size distribution of the feeds and fractions increments in the product. The results show that the mill is dedicated to small size of the feed. The specificity of work due to the size of grinding media and their strength does not allow the grinding of large particles, i.e. above 0.5 mm. The content of the 50-100 μm fraction in the grinding product of coarse feed amounts to 9.4%, while in the fine feed it is 11.2%. Fraction above 100 μm in the fine feed is 40%, while for the coarse feed it is about 44%. What's more, one must take into account that the separation of a narrow fraction and selection of optimal grinding conditions is an extremely difficult process. The generalization of the data to the one model described by the polynomial function did not give a good fit. Different sets of grinding media significantly influenced the increments of the particle size fractions. The mean square error of the fit was large and affected the inference process. Therefore, it was decided to present separate equations for individual grinding media sets. It is necessary to select other grinding parameters to obtain a higher yield of mentioned particle size fractions or to pre-separate it from the feed. However, if 0-50 μm particle size fraction is the most expected one, the proposed scheme research will be suitable for preparing the raw material for the production process of the lime sorbents. The results of the research are presented in Table 6.

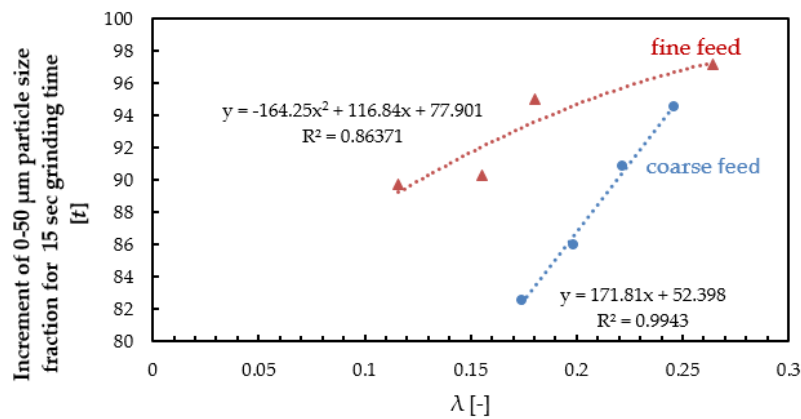


Fig. 5. Relation between λ values and increments of particle size fractions for fine and coarse feed

Table 6. Yield of 50-100 μm particle size fraction in the products of limestone grinding for fine and coarse feed

Feed size [mm]	Grinding media type (designation)	Yield of 50-100 μm particle size fraction at the specified grinding time					
		Time [s]					
		5	10	15	20	25	30
0-0.5	12/2 (1)	10.91	11.88	6.56	5.07	3.33	3.59
	12/1.5 (2)	11.83	10.57	6.19	3.05	2.36	0.51
	Mix (3)	12.95	9.72	3.38	1.40	0.48	0.35
	10/1 (4)	9.81	5.40	2.00	0.95	0.20	0.23
	12/2 (5)	10.24	11.76	7.86	7.58	5.58	3.04
0-1	12/1.5 (6)	10.58	10.81	10.22	4.43	2.18	2.42
	Mix (7)	9.73	8.51	6.67	2.17	1.08	1.19
	10/1 (8)	8.89	6.21	2.26	0.54	0.53	0.44

One-dimensional models of the 50-100 μm particle size yield were prepared using least squares method, as a result of which a functional dependence described by the third-degree polynomial (Eq. 3) was obtained. It can be concluded that the results of modelling are satisfactory (Table 7). Fig. 6 show

graphs illustrating the relationship between sorbent yield and grinding time for all types of grinding media used.

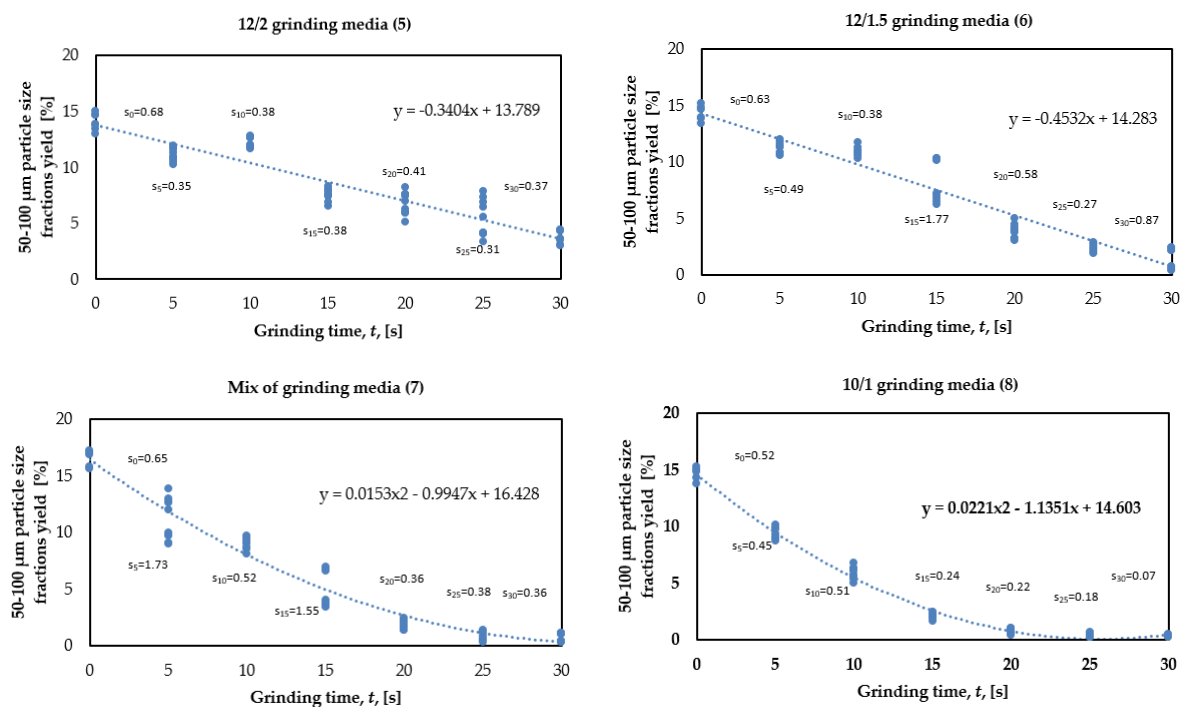


Fig. 6. Models of the 50-100 µm particle size fractions yield (axis y) determined for particular grinding media types (5), (6), (7), (8) depending on time (axis x) taking into account average mean values of fine and coarse feed

Table 7. Evaluation of the model fitting for 50-100 µm yield for average mean values of fine and coarse feed

Grinding media type	MSE [%]
12/2	1.318958
12/1.5	1.337333
mix	1.190856
10/1	0.424302

The value of the MSE index indicates very small differences between experimental and model data. It can be clearly seen that the error for the 50-100 µm particle size fractions yield model is on average 1.1%. In the majority of cases, it is expected that the yield of particle size fractions determined with the help of models will be characterized by an error not exceeding 1%.

The yield of the 0-50 and 50-100 µm size fractions is characterized by an opposite trend. More specifically, as the grinding time increases, the content of the 0-50 µm fraction in the final product increases, however, the 50-100 µm class yield is significantly reduced. It may be influenced by the 50-100 µm fraction content in the feed (9-11%) or the grinding conditions. The content of interesting particle size fractions in the feed is too small to undergo preliminary screening, which may be related to the necessity of introducing other preparatory operations or conditions for their conducting. The content of the fraction above 100 µm in the feeds is approximately 40%.

4. Conclusions

Sorbents production process for flue gas desulphurization is extremely important, especially due to the increasing production of air pollutants from various sources and long-term preventive actions. The production of such substances is associated with the necessity of obtaining fine limestone powder, which is their key ingredient. One of the possible ways is to use a grinding process that is characterized by high energy consumption. The introduction of electromagnetic mills becomes a good alternative that

allows the production of required materials while reducing energy costs. Therefore, the selection of optimal grinding conditions and the possibility of forecasting the effects of the process is a key issue. Based on the experiments carried out, models were obtained that highly reflect empirical data.

Coefficient of the exponential function is the only factor that affects changes in the shape of the model. The selection of 15 s grinding time was determined by the grinding kinetics characteristics of the raw material, according to which after this time further grinding was characterized by significantly lower increase efficiency of the analysed fraction. Longer grinding time provides only unnecessary energy expenditure with no effect on the material comminution. The feeds behaved similarly, which is why particle size distribution models for grinding product 0-50 μm are presented in one Fig. (models based on a quadratic equation). The grinding result was separated from each other for particle size fraction 0-50 and 50-100 μm , because in this case the phenomenon proceeded differently. While the second way of modelling was dedicated to the increase of the particle size fraction in grinding products due to different characteristics of empirical points distribution.

The largest increment of the 0-50 and 50-100 μm particle size fractions was noticed when using 10/1 grinding media.

The average fit was 96.5%, with the majority of results above 96%. The research allowed to determine the impact of changes in the parameters of the mill and the feed, which will allow to determine the controls for the system's continuous operation in the future. The paper focuses on the analysis and modelling of yields and increments of particle size fractions 0-50 and 50-100 μm , which are the key material for the production of limestone sorbents for the purification of exhaust gases. It can be observed that electromagnetic mill could be an efficient device to be used in limestone processing facilities when all stages of the development will be completed.

Acknowledgments

The research was performed on the installations co-financed by the National Centre for Research and Development, Poland, under Applied Research Program, project no. PBS3/B3/28/2015.

The research reported in this paper as well as the open access charge was co-financed by statutory activities AGH no. 11.11.100.276

References

- BERTHOUEX P.M., BROWN L.C., 2002. *Statistics for Environmental Engineers*, CRC Press LLC, Boca Ronto, FL, USA.
- BOND, F.C., 1952. *The third theory of comminution*. Trans. AIME, 193, 484–494.
- CAMERON I.; HANGOS K., 2001. *Process Modelling and Model Analysis*, Academic Press, London, UK, pp.100-249.
- CHODOROWSKI J.; SALOMOWICZ W., 2004. *Investigation of sorbent molecules*, The script SGSP, Warsaw, Poland. (in Polish)
- DELBONI, H.J.; MORRELL, S., 2002. *A load-interactive model for predicting the performance of autogenous and semi-autogenous mills*. KONA Powder Part. J., 20, 208–222.
- FOSZCZ D., 2006. *Estimation of regression function parameters by classical and bootstrap methods*. Górnictwo i Geoinżynieria, 30, 57–78. (In Polish)
- GUPTA, V.K.; SHARMA, S., 2014. *Analysis of ball mill grinding operation using mill power specific kinetic parameters*. Adv. Powder Technol., 25, 625–634.
- HLINCIK T.; BURYAN P., 2013. *Evaluation of limestones for the purposes of desulphurization during the fluid combustion of brown coal*. Fuel, 104, pp.208-215.
- KICK, F., 1885. *Das Gesetz der Proportionalen Widerstände Und Seine Anwendungen*; Felix: Leipzig, Germany.
- KING R.P., 2012. *Modeling and Simulation of mineral processing system*, Elsevier Science Ltd.: New York, NY, USA, pp. 30-170.
- KOTOWSKI C.; RATAJCZAK T., 2010. *The Carboniferous limestones from Czatkowice - their technological potential and possibilities of utilization*. Górnictwo i Geoinżynieria, 34, pp.339-348. (in Polish)
- KRAWCZYKOWSKI D.; FOSZCZ D., OGONOWSKI S.; GAWENDA T., WOŁOSIEWICZ-GŁĄB M., 2018. *Analysis of the working chamber size influence on the effectiveness of grinding in electromagnetic mill*. MEC 2018: Mineral Engineering Conference: Zawiercie, 26–29 September, pp. 43.
- LEE-ING T.; CHUNG-HO W., 2002. *Statistica V5.5 Basic Statistic Analysis*. TasnghHai Taiwan Publisher, Taiwan.

- MAXWELL S. E.; DELANEY H. D., 2004. *Designing experiments and analysing data: Model comparison perspective (2nd ed.)*. Mahwah NY: Lawrence Erlbaum Associates, New York, NY, USA.
- NAPIER-MUNN, T.; WILLS, B.A., 2006. *Wills' Mineral Processing Technology, 7th ed.*; Elsevier Science Ltd.: New York, NY, USA, pp.90–108.
- NIEDOBA T., 2013. *Multidimensional characteristics of random variables in description of grained materials and their separation processes*. Wydawnictwo Instytutu Gospodarki Surowcami Mineralnymi i Energią, Krakow, Poland, pp.127-139. (in Polish)
- NOLAN, PAUL S., 2000. *Flue Gas Desulfurization Technologies for Coal-Fired Power Plants*, The Babcock & Wilcox Company, U.S., presented by Michael X. Jiang at the Coal-Tech 2000 International Conference, November, Jakarta, Indonesia
- OGONOWSKI, S.; WOŁOSIEWICZ-GŁĄB, M.; OGONOWSKI, Z.; FOSZCZ, D.; PAWEŁCZYK, M., 2018. *Comparison of Wet and Dry Grinding in Electromagnetic Mill*. Minerals, 8, 138.
- OGONOWSKI, S.; OGONOWSKI, Z.; SWIERZY, M., 2017. *Power optimizing control of grinding process in electromagnetic mill*. In Proceedings of the 2017 21st International Conference on Process Control, Štrbské Pleso, Slovakia, 6–9 June; pp. 370–375.
- PAWEŁCZYK M., OGONOWSKI Z., OGONOWSKI S., FOSZCZ D., SARAMAK D., GAWENDA T., KRAWCZYKOWSKI D., 2015. *A method for dry grinding mill electromagnetic* (in polish), patent application no. P.413041 (06.07.2015).
- PETRAKIS E. AND KOMNITSAS K., 2017. *Improved modeling of the grinding process through the combined use of matrix and population balance models*. Minerals, 7 (5), 67.
- RITTINGER, P.R., 1867. *Lehrbuch der Aufbereitungskunde*; Ernst and Korn: Berlin, Germany.
- SHIN, H., LEE, S., JUNG, H.S., KIM, J-B., 2013. *Effect of ball size and powder loading on the milling efficiency of a laboratory-scale wet ball mill*. Ceram. Int., 39, 8963–8968.
- TOUIL, D.; BELAADI, S. FRANCES C., 2008. *The specific selection function effect on clinker grinding efficiency in a dry batch ball mill*. Int. J. Miner. Proc., 87, 141–145.
- TUMIDAJSKI T., 1997. *Stochastic analysis of properties of grained materials and their separation processes*, The script AGH, Krakow, Poland. (in Polish)
- TUMIDAJSKI T.; SARAMAK D., 2009. *Methods and models of mathematical statistics in the processing of mineral resources*, The script AGH, Krakow, Poland. (in Polish)
- WALKER, D.R.; SHAW, M.C., 1954. *A physical explanation of the empirical laws of comminution*. Trans. AIME, 199, 313–320.
- WANG Y.; FORSSBERG E., 2007. *Enhancement of energy efficiency for mechanical production of fine and ultra-fine particles in comminution*. China Particuology, 5, pp.193-201.
- WOŁOSIEWICZ-GŁĄB M.; PIĘTA P.; FOSZCZ D.; OGONOWSKI SZ.; NIEDOBA T., 2018. *Grinding Kinetics Adjustment of Copper Ore Grinding in an Innovative Electromagnetic Mill*. Applied Sciences, 8(8), 1322.