Characterization of Changes in Particle Size Distribution by the PaRMAC Evaluation Method†

S.P.E. Forsmo1, S-E Forsmo1
and P-O Samskog1,2
1 LKAB, Malmberget, Sweden
2 Luleå University of Technology, Sweden

Abstract

PaRMAC is a newly developed evaluation method for the particle size distribution in agglomeration. The particle number distribution is divided into three size categories: 1-10 µm ("sand"), 10-40 µm ("stones") and 40-100 µm ("block"). The "stones" form the basic framework of the pellets. The "sand" is needed to fill in the voids. The relationship between the size categories is quantified by comparing each smaller size to the next larger. Two ratio factors are obtained and used as x-y coordinates. The obtained point defines the status of the sample. By following changes in the position of this point, changes in the original size distribution can be traced. The PaRMAC method allows us to visualise and compare hundreds of samples at the same time.

Changes in the particle size distribution of LKAB iron ore concentrates are tracked as the agglomeration process advances. The magnetite concentrates begin to agglomerate as early as during filtration. The bentonite binder redisperses the agglomerates. The dispersion grade varies due to changes in the efficiency of the bentonite mixer.

1. Introduction

The pelletizing process of iron ore concentrates includes wet grinding of the concentrate to the desired fineness, addition of additives, filtration to about 9% humidity, addition of binder, balling to a mean pellet size of 11 mm, and finally drying and induration at high temperatures to achieve the desired mechanical strength and metallurgical properties. The iron ore mined at LKAB, Sweden, is magnetite which oxidises to hematite during induration. This oxidation is an exothermic reaction releasing a large amount of energy. As much as two-thirds of the energy used for pelleting comes from the magnetite oxidation to hematite. The particle size distribution affects the pellet properties in a complex manner because it not only changes the porosity and packing properties, but also the oxidation pattern and the temperature profile in the pellet bed during induration.

The packing properties of dry powders have been widely studied [1,2]. A comprehensive review regarding both experimental work and mathematical models in this area was given by Yu and Zou [3] in the previous volume of this journal. The target when optimising the agglomeration properties of magnetite concentrates is not necessarily a high packing density as often seems to be the case in the area of packing dry powders. The pellet porosity should be a compromise between the pellet's mechanical strength and an open inner structure for better gas transfer during oxidation and reduction.

Traditionally, the particle size distribution of iron ore concentrate is measured by screening (weight distribution) or by laser diffraction methods (volume distribution). Also, the specific pressure drop through a packed bed of particles (Blaine index or Svensson surface) is commonly used to describe the fineness of materials. When this agglomeration research was started, we knew from practical experience that none of these methods successfully predicts the pelleting process behaviour or the pellet quality as a function of the particle size distribution.

The formation of a green pellet can be regarded as masonry work on a micro scale. A specific number of particles with suitable size ratios is needed to do the work, a strong and at the same time open green pellet. Upon looking for a measuring instrument that could measure the size distribution of the number of...
particles instead of weight or volume distributions, the scanning laser microscope was found.

Another difficulty that was faced was to evaluate changes in the particle size distribution when a large number of samples is to be considered. Visual inspection of the size distribution curves can cope with maybe five to ten samples at a time. Numerical inspection of the curves by using, for example, \( k_{90} \) values reduces the inspection to only one, or a few points on the size distribution curve, and information on crossing curves is easily lost. A better evaluation method was needed and the PaRMAC method was developed. The name PaRMAC stands for Particle Ratios Method for Agglomeration Characterization.

With the PaRMAC method, the suitability of a particle mixture as masonry raw material is described by dividing the size distribution curve into three size categories. They are ratioed to each other and the ratio factors are used as \( xy \) coordinates. This point describes the original particle size distribution of the sample. The PaRMAC method allows us to inspect and compare hundreds of samples at the same time and to visualise the differences in a simple and practical manner. PaRMAC has proved to be a good help in understanding how to accomplish desired changes in the size distribution. In this article, the PaRMAC method is described in detail and some practical applications of the method are given.

2. Measuring methods

The particle size distribution is measured using the Partec 100 instrument by Lasentec. It uses scanning laser microscope technology. The laser beam is focussed to a small beam spot with a high intensity at the focal point. The laser beam scans at a high and constant velocity, and the time it takes for the beam to scan across a particular particle is a direct measure of the particle size.

The Partec 100 instrument measures a random chord across the particle. The iron ore particles have shapes which are complex and irregular. As they are often somewhat needle-shaped, they show fairly short chord values more frequently. The additive particles, olivine and dolomite, are quite round and show about the same chord value in all directions. Depending on this difference in particle shape, screening, PaRMAC and laser diffraction all give different results. The magnetite concentrate and the additives are ground to the same fineness measured by sieving (% <45 \( \mu m \)). PaRMAC describes the additive particles as large compared to magnetite. In the Malvern diffraction pattern measurement, the additive particles are small and practically disappear under changes in the large magnetite particles [4]. This discrepancy between different measuring methods is a well-known problem when measuring the size distribution of particle mixtures with different particle shapes.

The particle number distribution measured by the Partec 100 can be mathematically recalculated to mass and volume distributions. At present, however, the raw data from the measurements are used. Possibly, in future, calculation of the volume distribution could be performed as a complement to the particle number distribution in order to collect additional information about changes in the large particles. This applies especially to the new digital instrument versions, where the counting time can be increased to also obtain good reproducibility for the largest particles.

The measuring range of 1.9 to 1000 \( \mu m \) is divided into 37 measuring channels, of which about 30 are active when measuring iron ore concentrates.

The Partec 100 measurements are made in a slurry containing 50 wt-% solids under high-shear mixing conditions. Moist samples such as filter cakes and green pellets can be measured directly without drying. The binding agent, bentonite, is invisible to the PaRMAC when it is added in amounts of 0.5 weight\%. Therefore, even green pellets can be measured. The possibility of measuring the particle size distribution in the natural process water environment, without drying and redispersion, has proved to be especially valuable.

Maintaining a constant measuring geometry, mixing speed and slurry solids content are prerequisites for a successful measurement. Reproducible results also require special care in sampling, sample preparation and dispersion.

3. The PaRMAC evaluation method

PaRMAC stands for Particle Ratios Method for Agglomeration Characterization. It was published in the proceedings of the XXth IMPC conference in Aachen, 1997 [4]. Our aim is to use the PaRMAC method in correlating and optimising the particle size distribution in pellet feed with respect to the pellet properties. The same principle of ratioing desired size fractions can, of course, be used in other applications as well as for mass and volume distributions.

With the PaRMAC method, the particle mixture is considered as a ternary particle system. The size classes are chosen empirically to describe the different behaviour of the particles.
The three size categories were selected and designated as follows:
- “sand” about 1-10 μm
- “stone” about 10-40 μm
- “block” about 40-100 μm

The designations sand, stone and block were chosen to describe the use of these different fractions as building materials for the green pellets. The “stones” form the basic framework in the pellet, the “sand” is needed to fill in the holes between the stones. The “blocks” are baked inside the green pellet with enough of both sand and stone. The names simplify discussions over changes in the size distributions. Also, they associate the complicated science of balling green pellets to the art of making sand cakes, well known by all small children. A firm sand cake can be created by mixing the three components in varying proportions and by adding a suitable amount of water.

The “sand” category was chosen to describe the particles that show nearly colloidal behaviour and have a comparably large surface area. Therefore, this fraction is expected to be a good indicator of surface chemical reactions such as flocculation, dispersion and topochemical oxidation. The measuring range of the Partec 100 instrument sets the lower limit of the sand fraction at about 1 μm. This was considered acceptable from the agglomeration point of view, because the average pore size in green pellets is around 1-2 μm.

LKAB produces pellets at both their Malmberget and Kiruna operations. For the Kiruna pellet concentrates, most of the particles are between 10 and 40 μm in size. The upper limit of 40 μm is set by the cut-off size of the hydro-cyclones in closed-circuit grinding. This size range was chosen for the category designated as “stones”.

The upper limit for the “block” designation was chosen to be about 100 μm because in pellet concentrates, particles larger than this are few and of minor interest. The blocks probably show characteristic oxidation behaviour, because the oxygen needs to diffuse longer distances inside the particles.

The relationship between these three size categories is quantified by comparing each smaller size category to the next-larger size category. The following ratios are calculated:

\[
\text{sand factor (x)} = \left(\frac{\% \text{ sand}}{\% \text{ stone}}\right) \times 100 - 100 \\
\text{stone factor (y)} = \left(\frac{\% \text{ stone}}{\% \text{ block}}\right) \times 100 - 600
\]

The constants of 100 and 600 have been chosen so that the normal values for the Malmberget pellet feed lie near the origin.

These two factors are used as x and y coordinates and the resulting (x,y) point describes the overall size status of the sample in the PaRMAC chart. Changes in the position of this point describe changes in the original size distribution curve. Typical directions of changes are shown in Figure 1. In open circuit-grinding, the PaRMAC point moves towards higher positive x and y values as the material becomes finer. Cycloning lifts, typically, the PaRMAC point upward towards higher stone factor values. The exact direction of the change depends on the cut-off size of the hydro-cyclones.

The changes in the slope of the cumulative particle size distribution curve can also be interpreted from the PaRMAC chart. The steeper the curve, the further up and to the left the point moves. The particle size distribution becomes more “monosize” and obviously, more difficult to agglomerate.

The precision (defined as 2σ) is ±0.5 units for the sand factor (x) and ±10 units for the stone factor (y). The chosen limits for the three different size categories have been successful for our application.

4. Applications of the PaRMAC evaluation method

4.1 A comparison of pellet concentrates from different LKAB concentrating plants

A set-up of magnetite pellet concentrates from different LKAB concentrating plants is shown in the PaRMAC chart in Figure 2. The quality variations for different concentrates do not describe comparable process quality variations because of different sampling intervals.

The PaRMAC points for the Kiruna and Svappavaara pellet concentrates (KPC and SPC) lie at lower sand factor (x) values and higher stone factor (y) values.
compared with the Malmberget pellet concentrate (MPC). This is due to closed-circuit grinding in Kiruna and Svappavaara. In Malmberget, we have open-circuit grinding in three steps. The KPC and SPC are much finer by screening compared with the MPC (KPC 84% and MPC 68% <45 μm, respectively). However, the size distribution curve for KPC is much steeper due to cycloning. The proportion of “sand” to “stone” decreases and “stone” to “block” increases, the distribution becomes more monosize.

A more monosize distribution should result in higher porosity in the green pellets. This is indeed the case. The porosity in Kiruna and Svappavaara green pellets is typically somewhat higher than in the Malmberget pellets (32% and 30%, respectively).

The differences in the particle size distribution between KPC and MPC can also be seen in the oxidation pattern of these materials. Typical thermogravimetric analysis (TG) results for KPC and MPC are shown in Figure 3. MPC with higher PaRMAC x-values oxidises at low temperatures (<400°C) more to maghemite than KPC. This low-temperature oxidation of magnetite to maghemite is generally agreed to be topochemical in the literature. KPC with higher PaRMAC y-values gains in the oxidation rate when the large number of medium-sized particles is oxidised.

4.2 The benefit of visualising a large number of near-similar samples in the PaRMAC chart

In our earlier publication [4], an example was given regarding the visualisation of a large number of samples at the same time in the PaRMAC chart. This is shown in Figure 4.

In Malmberget, three open-circuit grinding sections were used to produce the pellet concentrate, MPC.

The control value for all three sections was 68% <45 μm by sieving. The PaRMAC points for samples from section 3 differ significantly to those from sections 4 and 5 although they all showed similar values by sieving. The reason is a flow of screened oversize particles connected to the primary mill in section 3.

In Table 1 screen analysis, Malvern and PaRMAC values are given for two sets of samples from these three sections. When sampling, the control values were 66% (section 3) and 64% (sections 4 and 5) <45 μm by sieving. The table describes six samples altogether. It demonstrates the difficulty of comparing particle size distributions expressed in figures, especially if the curves cross each other. The PaRMAC chart in Figure 4 shows 326 samples altogether. By only glancing at the PaRMAC chart, one can directly point out the samples that differ.
Table 1: Sieve analysis, Malvern and PaRMAC values for Mahnberget pellet concentrates (MPC) produced in three parallel production sections.

| Screening | Malvern K<sub>90</sub>/µm | Malvern K<sub>50</sub>/µm | Malvern K<sub>10</sub>/µm | PaRMAC (x, y) |
|-----------|----------------|----------------|----------------|--------------|
| Sample pair 1: |
| Section 3 | 66.3 | 5.2 | 46 | 138 +0.7, +59 |
| Section 4 | 63.5 | 5 | 51 | 175 +7.5, +80 |
| Section 5 | 63.9 | 5.1 | 51 | 162 +5.3, +70 |
| Sample pair 2: |
| Section 3 | 67.5 | 4.4 | 47 | 173 +0.5, +59 |
| Section 4 | 61.8 | 4.6 | 48 | 162 +5.2, +59 |
| Section 5 | 63.5 | 4.4 | 42 | 134 +6.2, +72 |

4.3 Particle size distribution in real process conditions interpreted by PaRMAC

Because the measurements are performed on a slurry with 50 weight-% solids, moist samples such as filter cakes and green pellets can be measured directly without drying. This allows us to study the particle size distribution and dispersion in real process conditions. In the case of green pellets which contain the bentonite binder, it is in general not possible to measure the particle size distribution at all, if the pellets need to be dried first. The bentonite binds the dried particles together so hard that redispersion is not possible.

The PaRMAC charts in Figure 5 compare the dried pellet concentrates, the moist and dry filter cakes and the green pellets at the concentrating and pelletising plants of LKAB located in Mahnberget (MPC), two plants in Kiruna (KPC-KA1 and KPC-KA2) and in Swappavaara (SPC). The pellet concentrates contain only magnetite, the filter cakes contain magnetite and olivine, and the green pellets contain magnetite, olivine and bentonite. The individual sample sets do not represent contemporary material flows. However, the PaRMAC charts give an interesting picture of how the particle size distribution changes due to different unit operations and different process design.

The pellet concentrates mixed with olivine are stored in large slurry buffer tanks before filtration. Therefore, the filter cakes show less variation in the PaRMAC co-ordinates than the pellet concentrates.

Fig. 5 PaRMAC chart on (○) dried pellet concentrates, (□) moist filter cakes, (△) dry filter cakes and (◇) green pellets at LKAB Mahnberget, Kiruna (2 plants) and Swappavaara.
This can be seen in Figure 5a, where the number of samples is large enough to describe real process variations.

The addition of olivine to the magnetite concentrate changes the PaRMAC coordinates. According to laboratory tests, addition of 3.6% olivine to the pellet concentrate slurry results in 10-20 units lower PaRMAC y-values. Therefore, the filtered pellet feeds should lie at somewhat lower y-values than the magnetite concentrates. This is the case for the dried filter cake samples. However, the moist filter cake samples measured directly without drying lie at much lower x- and y-values. This applies to all plants in varying degrees. This difference in the coordinates between the flotated and unflotated concentrates can also be seen visually during sample preparation. However, the moist filter cake samples measured directly without drying lie at much lower x- and y-values. This applies to all plants in varying degrees. This difference in the PaRMAC coordinates of humid and dried filter cake samples is explained by agglomeration during filtration. These agglomerates, which do not contain any bentonite binder, need to be dispersed before balling. Otherwise, it would lead to problems in pellet quality.

The addition of bentonite disperses the agglomerates formed during filtration. However, the degree of redispersion depends on the strength of the agglomerates, the effectiveness of the bentonite mixer, and also on the energy input in the balling circuits.

In Malmberget, we had the opportunity to follow this redispersion pattern when the old bentonite mixer in the pellet plant was changed to a new Eirich mixer [5]. After the new mixer was installed, the green pellet coordinates in the PaRMAC chart were shifted further to the right towards higher sand factor (x) values, indicating more effective dispersion in the bentonite mixer, see Figure 6.

The flotated pellet concentrate in Kiruna KPC-KA2 shows less agglomeration due to filtration than the unflotated concentrate KPC-KA1, see Figures 5b and 5c. This can be explained by flotation reagent adsorption on magnetite surfaces. This difference in surface characteristics between the flotated and unflotated concentrates can also be seen visually during sample division. The moist, flotated pellet feed is more freeflowing and shows less tendency to agglomeration.

The PaRMAC measurements on samples containing bentonite are carried out by diluting the samples to 50 weight-% solids by the process water. When parallel samples of green pellets were prepared using a combination of process water and distilled water, the PaRMAC coordinates in clean water were systematically lower in both x- and y-values, indicating a lower degree of dispersion, see Figure 7. This could be either due to higher pH or the high salt content in the process water. The pH and the salt content in the process water show periodical variations, and the water quality is also different at different plants [6]. These variations might not be of great interest from the agglomeration point of view in the production scale, but they do play a role in the precision of the PaRMAC laboratory measurements. Therefore, samples containing bentonite are always diluted with filtered process water. For samples without bentonite, the quality of the water used in sample preparation is irrelevant.

This example shows that the concept of particle size distribution in pellet feeds is somewhat ambiguous. The question that arises is whether the pellet properties should be correlated to the primary particle distribution or to the actual size distribution of agglomerates found in real process conditions. The oxidation rate in single particles could be expected to correlate with the fineness of primary particles measured in dried concentrates. The packing properties and the oxidation behaviour inside a single pellet
could, however, also depend on the material’s tendency to agglomerate during filtration, on bentonite mixing efficiency, and on the dimensions of the balling drums. These phenomena are difficult to simulate on a small scale. They need to be considered when results from small-scale studies are scaled up to the production scale.

5. Discussion

The examples given above show the versatility of the PaRMAC method. It is a great help for a scientist to be able to view a large number of size distributions on a single chart and to be able to rapidly obtain an impression of deviating samples or sample groups.

The use of size distributions based on the number of particles has given more information about the behaviour of those size fractions which are significant for the pelletising process. Most of the unit operations in the concentrating plant lead to varying degrees of skewness in the size distribution curves. Changes in the process lay-out at the early stages of the concentrating process have been shown to affect the size distribution in the final pellet concentrate. Combined with process knowledge, the PaRMAC chart will show both optimal working areas and areas to be avoided.

The use of proportions between size fractions instead of absolute particle sizes seems to give a fair correlation to the pellet porosity and also to the magnetite oxidation behaviour. We are intensively studying this further.

The possibility of measuring the particle size distribution in real process conditions has shown that although the distribution of primary particles would be constant, variations in aggregate size distribution could occur because of variations in bentonite mixing efficiency or changes in balling circuits. These variations have previously not been detectable.

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Author's short biography

Seija Forsmo
Seija Forsmo works as a senior research engineer in LKAB. She works presently to develop new analytical measuring methods for the characterization of the agglomeration properties of iron ores.

Sten-Evert Forsmo
Sten-Evert Forsmo works as a senior research engineer in LKAB. He works presently to correlate raw material properties to the pelletizing process and to the pellet quality.

Per-Olof Samskog
Per-Olof Samskog received his Ph.D. physical chemistry at the University of Stockholm 1979 and spent two years as visiting scientist at the University of Alabama US. He is currently manager of Systems Technology at the Swedish mining company LKAB and adjunct Professor at the Luleå University of Technology, Sweden.