X-RAY MICROTOMOGRAPHY FOR SEQUENTIAL IMAGING AND ANALYSIS OF IRON ORE PELLETS UNDER REDUCTION

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Abstract

This work has been a part of the development of a method where x-ray microtomography and image analysis are used to quantify the crack distribution and propagation in iron ore pellets during reduction. Reduction experiments on 7 pellets, were performed in 4 steps during the transformation from hematite to magnetite at 500 °C. The pellet microstructure is imaged and visualized at each step using x-ray microtomography and the crack distribution characterized with image analysis.

For the chosen pellet composition, networks of cracks are already visible in the unreduced state and show a successive growth during the first 30 minutes of reduction. As the transformation to magnetite is complete, crack growth has stopped and in several cases a partial regression is observed.

It can be concluded that the use of X-ray microtomography has the potential to become an important tool for material characterization in iron ore pellet research due to its ability to gather information from the bulk of the material at several different stages of a process.
1. Introduction

Hematite will when exposed to a reducing atmosphere at temperatures around 500°C undergo a phase transformation to magnetite. This will lead to a volume swelling of the microstructure that may reduce the material strength, due to increased internal stress. The degree of disintegration during the transformation to magnetite is used as a pellet quality parameter during pellet production (low temperature disintegration test, or LTD, according to ISO13930). Test conditions are designed to simulate the conditions in the upper part of a blast furnace, and the strain caused by the phase transformation is combined with a mechanical strain, as 0.5 kg pellets are tumbled in a rotating furnace for one hour.

Previous observations have however indicated that results from these laboratory tests not always correlate with the disintegration properties of the corresponding material in the blast furnace. The LKAB olivine pellets for example have fairly high values of disintegration during laboratory testing. Probe samples taken from the LKAB experimental blast furnace [1] has however shown that disintegration rates within the furnace are considerably lower and outstanding pellet performance have been achieved despite the level of disintegration at laboratory scale [2].

Lack of quantified data have so far made correlations between microstructural effects, experimental conditions and resulting pellet properties difficult. In order to describe how factors such as temperature, diffusion, gas composition, permeability and pellet strength affect the disintegration, there is a need to characterize how initial weaknesses grow into larger cracks leading to the breakdown of the pellet. This is expected to lead to a deeper understanding of when and to what extent, variations in pellet quality at laboratory scale can be expected to affect blast furnace performance.

The aim with the current study is to investigate the possibility of using X-ray microtomography (XMT) and image analysis for quantitative analysis of cracks within iron ore pellets under reduction. XMT is a non-destructive imaging technique that allows repeated imaging of the full three dimensional material microstructure, coded in density [3]. Hence it is possible to image and analyse the same samples at different stages of a certain process, and follow the material response due to thermal, chemical and mechanical loads [4,5].

Similar methodology has for example been used for studies of metal powder compacts during sintering [6] and for analysis of the crack propagation in cast iron due to fatigue [7].

Moreover, the fact that XMT gives a three dimensional representation of the material makes it a beneficial tool for imaging and analysis of cracks, which due to their lack of isotropy and symmetry are hard to predict and model based on two-dimensional image data. The technique is non-destructive and no preparation of the samples is required before examination, which in itself might introduce new defects or cause propagation of already existing cracks.

The use of 3D image analysis makes it possible to identify the cracks inside the studied material, and make a quantitative characterization of these features [8-11].

On relevant materials for this study, this type of textural analysis have been used for studies of pore structure and porosity in iron ore sinter [12,13], and iron ore pellets [14].

2. Material and methods

2.1 Material properties and processing

Raw material and pelletization

A magnetite concentrate was used, previously identified as a cause for increased deterioration rates during reduction due to its distribution of magnesium within the magnetite lattice. This
choice was made to assure that the deterioration of the material would be sufficient for the investigation. The concentrates were mixed with additives (olivine, quartzite, burnt lime and bentonite) and balled on a rotary disc to pellets with sizes of 10-12 mm. Pellets were loaded and indurated in a shaft furnace with a batch size of 40 kg. The maximum temperature during induration was 1300 °C.

Reduction and sample setup
The reduction experiments were performed in a laboratory furnace, with a constant temperature of 500 °C, a gas composition of 20 % CO, 20 % CO₂, 60 % N₂, and a gas flow of 5 L/min (0 °C, 1 atm). The reduction experiments were performed stepwise and interrupted after 10, 20, 30 and 60 minutes, as shown in Figure 1. The investigated pellets were scanned with X-ray microtomography before reduction, and after each subsequent stop. Heating (90 min) and cooling (30 min) were performed in a pure nitrogen atmosphere. The reduction degree was monitored by measuring the weight of the pellet samples during the experiments. There was no indication that any reaction took place during the heating and cooling cycles.

2.2 X-ray microtomography
The scans were all carried out using the X-ray microtomography system shown in Figure 2. The system includes a microfocus x-ray source (L7901-01) and detector unit (C7876-10) from Hamamatsu Photonics.

The iron ore pellet samples are positioned on a motorized rotation stage (Linos Photonics, RT120 ST), which together with the image acquisition are controlled from a host computer. During a scan 744 x-ray projection images were collected at equal angles as the sample makes one full rotation. The x-ray tube voltage and current were held at 70 kV and 90 µA, respectively. The source-sample and source-detector distances were 8.9 mm and 31.6 mm, respectively, resulting in a 3.6 times magnification and a theoretical pixel size of 30 µm/pixel. The dimensions of the projections were 640 x 509 pixels and the acquisition time 2.56s.
The tomographic reconstruction was carried with a cone-beam back projection algorithm (Feldkamp type) [15]. The results describe the density of the material, where the different constituents in the iron ore pellet are represented by different grayscale levels. Regions with high density like for example iron oxide particles appear bright while low density regions like vacant spaces, such as cracks and pores, will appear dark. Prior to and after reconstruction a number of artefact correction routines were applied to the data to remove ring artefacts and correct for beam hardening and geometrical misalignment [16]. The surfaces of the reconstructed pellet samples contained severe steak artefacts due to beam hardening. The outermost region in each pellet (thickness: 1.0 mm) was therefore extracted before analysis.

2.3 3D image analysis

The microstructure of the reconstructed samples was analysed using 3D image processing and analysis. Here the methodology is described briefly. For a more thorough description please consult [17].

The main focus was on identification and characterization of the crack network within the material. First the 3D image data from XMT is binarized by thresholding the grayscale information (histogram). This results in a binary image that only consists of two phases, and separates the dense iron ore material (mainly iron oxide) from the features that consist air, such as cracks and pores. After a subtraction of the background, only features such as pores and cracks are visible as white structures in an otherwise black 3D space. Image processing routines were used to separate more elongated features from features with close to spherical shape, and hence distinguish the cracks from the pores and bubbles. The software was developed in Matlab using the Image processing toolbox.

Next, all of the cracks were identified and labelled, based on the estimated crack length (CL), which here is taken as the Euclidean norm of the smallest box (aligned with the global coordinate system) that entirely encloses the feature, as shown in Figure 3a and b. Thus, given a bounding box with the sides $x_c$, $y_c$ and $z_c$, as shown in Figure 3b, the estimated crack length will be,

$$CL = \sqrt{x_c^2 + y_c^2 + z_c^2}$$

The global parameterization of the cracks in the pellets is carried out using spherical coordinates, as shown in Figure 3a. The origin in this coordinate system corresponds to the

![Figure 3](image)

Figure 3. The parameterization of cracks in the iron ore pellets is carried out using a spherical coordinate system (a). The length of a crack is obtained from the Euclidian distance of the smallest box that entirely encloses the crack (b). The radial analysis is carried out in spherical shells, with the width 0.5mm. The volume of these shells increase with radial distance (c).
estimated geometrical centre of the iron ore pellet, which is obtained from a centre-of-mass calculation, under the assumption that the pellet is a homogeneous solid. Apart from CL, the cracks are also characterised through their volume, which is calculated directly from the crack, and not from a bounding box (Figure 3b). It may therefore be considered as a more accurate size measure than CL. However it holds no information about the extension of the crack, as CL does, and might be hard to correlate to material strength. Using the parameterization from Figure 3a, a radial analysis is carried out, where the volume of the cracks is measured as function of the radial distance from the pellet centre. The analysis is carried out using a set of spherical shells, of width 0.5 mm, which runs in the radial direction from the pellet centre and 6.0 mm out, as shown in Figure 3c. The crack volume within each of these shells is measured and the crack volume fraction is obtained by division by the volume of the corresponding shell, which varies with the radial distance, as shown in Figure 3c.

3. Results and discussion

3.1 Visualization of the crack network

Figure 4a shows a center cross-section of one of the investigated iron ore pellets (p2) for different times of reduction and Figure 4b shows a visualization of the 3D crack network in the same pellet. Here, the four largest cracks (highest CL) are visualized in different color, in descending order: red, yellow, green and blue. It contains a relative large crack already before the reduction process has started. From the subsequent scans one can see a certain growth of the crack, especially in the scans collected after 20 and 30 minutes. However, after 60 minutes the crack seems to have reduced in size. This behavior can also be seen in the 3D visualizations (Figure 4b), and correlates well with the crack volume measurements (CV), obtained from quantitative analysis.

Figure 4. Reconstructed pellet structure (a) and 3D crack network (b) for different times of reduction. The crack volume (CV) for the largest crack, obtained from quantitative analysis, is given below the 3D visualization. The crack size increases successively during the initial 30 minutes. After the following 30 minutes the crack growth has stopped and a regression in size is observed.
3.2 Quantification of the crack network

Size measurements

Figure 5a-c shows the quantified information of the crack propagation in the 7 analysed samples (p1-p7), which in general all correlate with the visual interpretation made from Figure 4.

In average (black curve) the crack volume expands approximately 50% (from 10.0 mm$^3$ to 15.2 mm$^3$) during the initial 30 minutes. Between 30 and 60 minutes a regression of the crack volume occurs and the final size is in average approximately 10% larger (11.0 mm$^3$) than in the initial state. The crack shown in Figure 4 is here given by the p2-curve, with a volume variation close to the average. In two samples (p5 and p7) the crack volume remains more or less constant after the initial growth.

Figure 5c shows the estimated crack length according to Figure 3b and Equation 1. This measure displays the same relative changes over time as the measurement of the crack volume. In average, the increase in crack length is approximately 17% after 30 minutes reduction, and 3% after 60 minutes. Normalization is carried out with respect to the maximum value of CL that is obtainable from Equation 1, which corresponds to a (hypothetical) crack that span the full diameter of a 12 mm pellet in all three dimensions ($CL_{max} = \frac{3}{4} \cdot 12$).

It is reasonable to assume that crack length estimation is strongly coupled to the volume of the crack. It also seems probable that the actual length of the crack remains more or less constant after 30 minutes of reduction and that the decrease that is observed is caused by the fact that the width in some parts of the crack is reduced beyond the resolution of the method.

![Figure 5. Crack volume (a), Relative crack volume (b) and Normalized crack length as function of time in the reduction furnace, for 7 individual pellets (p1-p7).](image-url)
**Radial analysis**

Several different aspects and disturbances during pellet production can lead to the formation of cracks and crack morphology can be expected to differ depending on its original cause. Theories exist concerning how crack morphology affect crack propagation and pellet disintegration during reduction but these are hard to verify. For this purpose we have attempted to characterize the spatial distribution of the cracks. Figure 6a and b show 3D visualizations of the cracks within pellet p4 and p5, respectively, before reduction. In Figure 6a, the largest crack (in red) may be categorized as a radial crack with three radial streaks, at approximately equal angles. In Figure 6b we see a large concentric crack (red) and a much smaller radial crack in the center (yellow).

Figure 6c and d show the crack volume, in each reduction state, as function of radial distance from the center of the pellet, for the two samples shown in Figure 6a and b, respectively. The spherical shells in which the measurements are carried out (described in section 2.3) are...
very small close to the center of the pellet, as shown in Figure 3c. This sets the upper limit for the crack volume measurements and explains the small values that are observed in both Figure 6c and d, for radial distances close to zero. Moving from the center and out, the distribution in Figure 6c has a more rapid increase than the distribution in (d) does. This seems reasonable since the major part of the crack in Figure 6a is located close to the center of the pellet. Based solely on the distribution in Figure 6d the major part of the crack should be located in the region 2-5 mm from the pellet center. This seems to agree well with the shape and location of the crack in Figure 6a.

Figure 6e and f show the crack volume fraction with increasing radial distance, based on the results in Figure 6c and d, respectively. Here, the location of the peaks allows us to quantitatively distinguish between the radial crack (peak close to the center of the pellet) and the concentric cracks (peak at radial distance 2.75 mm). Although Figure 6e and f might give a more unambiguous description of the crack geometry itself, the volume distributions in Figure 6c and d might provide valuable information about how the crack geometry evolves during the reduction process.

4. Summary and conclusions

With the equipment and proposed experimental techniques it has been possible to characterize the crack network in pellets under reduction. The possibility to follow individual pellets through the process enables correlations between crack volume, experimental conditions and resulting pellet properties.

For the chosen pellet composition, networks of cracks are already visible in the unreduced state. Measurements of the crack volume show an almost linear growth during the initial 30 minutes. After 60 minutes the growth has stopped and in the major part of the samples a regression in crack volume is observed. In average, the increase in volume is +50% after 30 minutes and +10% after 60 minutes, compared to the initial non-reduced state.

A regression is also observed in the measurements of crack length. It is assumed to be caused by the fact that the width in some parts of the crack is reduced beyond the resolution of the method. The present method for parameterization of the crack length is coarse, and it is probable that the measurements of crack length also depends on their width (and hence the volume).

Radial analysis makes it possible to characterize and distinguish between radial and concentric crack geometries in order to study their impact on crack propagation and material strength. In future studies, attempts will be made to increase the spatial resolution and improve parameterization of the crack length, which would increase the accuracy of the method.

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6. References


