Toner Particle Shape Characterization by FE-SEM Image Analysis

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Abstract
Particle shape is becoming an increasingly important element in toner development. Controlling the particle shape provides an additional method for manipulating the fluidity, chargeability and transfer efficiency of toner. Improper flow and charging conditions can inhibit print performance due to excess or insufficient toner present at the toner addition mechanism and variability of adhesion to the photoreceptor surface. Previous methods of determining particle shape are shown to have insufficient resolution for recent and more technologically advanced toners due to decreasing particle size. A new method is presented utilizing the strengths of scanning electron microscopy for determining aspects of particle shape with a higher resolution and better accuracy for particles below 7 microns.

Introduction
Current trends in the dry toner industry are requiring decreasingly smaller particle sizes. Toner has seen a drop in average particle size from 16µm to less than 7µm over the past 15 years. Higher printing speeds require a smaller particle that is able to fuse and melt more quickly than a large particle is capable of. In addition, smaller particles more accurately attract to the surface of the photoreceptor producing a higher quality print.

Particle shape is a major factor in the flowability of a powder. Studies have shown a direct relationship between the shape and aspect ratios of particles and their rate of flow [1]. The invention of polyermized toners allows for the production of particles that are nearly perfect spheres. These particles have high circularity indices and have been shown to flow exceptionally well.

The typical methods of testing employ shape characterization based on the principles of flow cytometry. A sample of toner is suspended in solution, usually with the aid of a surfactant, and then is passed through an optical cell. A CCD camera with magnifying optics collects photos of the particles as they pass through the cell at a high frame rate. The digital images are then subjected to calculations to determine the circularity index (CI) for each particle.

The problem with this method arises from the low magnification and poor resolution of the CCD camera. The typical magnifications for an instrument of this type are 5X for a low-power, wide-area field, and 20X for a high-power, narrow-area field. The small size of toner particles requires the 20X magnification in order to resolve a single particle. The low resolution CCD sensor employed in these instruments prohibits the ability to accurately obtain images of toner particles. Currently, the lowest detection limit of a flow cytometry instrument is 0.7µm. Therefore, for a modern 7.0µm toner particle, a shape change of 10% is required to significantly affect the CI measurement. Attempting to determine a shape difference in a modern polymerized toner is not possible due to the very small differences in particle shape.

By relying on an optical method, the maximum magnification is an inherent limitation in this type of instrument. The wavelength of visible light effectively prevents magnification beyond 1000X. Scanning electron microscopy does not have this limitation. By collecting images at high magnification and high resolution, an accurate measurement of the CI can be made.

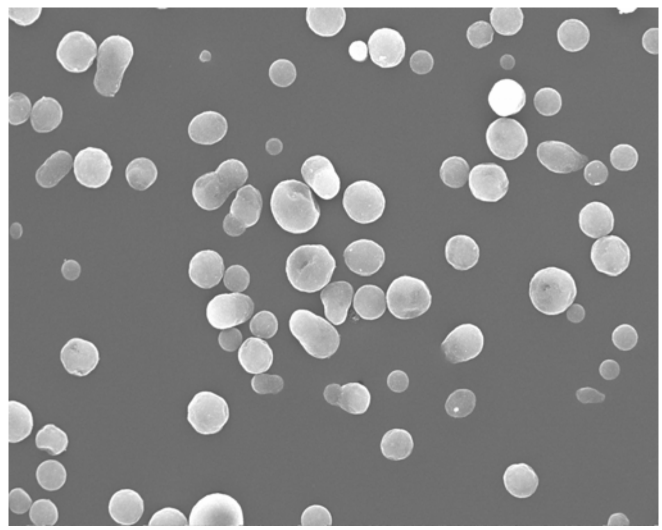

Figure 1. *A typical FE-SEM image of polymerized toner*

The FE-SEM Shape Analysis Method
The most important aspect of any image analysis method is the quality of the source image. FE-SEM image quality is dependent on the sample preparation and the conditions of the microscope. To prepare a toner sample for shape analysis, the toner particles are held onto SEM sample mounts with double-sided adhesive tape. The samples are then coated in a JEOL MFC-1300 platinum sputtering coater for 170 seconds.

The optimal microscope conditions for this analysis are done at a high accelerating voltage. This enhances the contrast around the outer edge of the particle and enables easy resolution between two closely spaced particles. The images are collected using a JEOL JSM-6345F FE-SEM. The accelerating voltage is 20kV at a beam current of 12µA. Although the magnification can be adjusted for higher resolution if necessary, it was found that images taken at 1000X using a 40 second image acquisition in photo mode provided the best conditions considering particle quantity and resolution. At least 150 particles should be analyzed to ensure sufficient accuracy. Six images are collected per sample. The collected images are 8-bit grayscale images with a resolution of 1280x1024.

Image analysis relies on applying mathematical calculations to an image in order to derive quantitative information about the contents of the image. Many of the algorithms and filters used during this image analysis are well documented and have been described elsewhere [2]. There are numerous software suites designed to handle image processing; Scandium v5.0 build 1066 by Soft Imaging Systems will be used here.

Preprocessing of the image is limited to a single iteration of a median filter. This is done to remove noise that is intrinsic in SEM images. Contrast optimizations are not done so that the true data captured in the image is preserved.

To analyze the image, a Separator filter is first defined. This filter causes a one pixel width line to be drawn around the perimeter of a toner particle. This separates many particles that would otherwise be detected as a single particle. To define the boundary shape, a step profile should be used with the dark background in the image. This is superior to the dark boundary shape which has a tendency to draw the line through particles. The Fine / Coarse setting should be adjusted manually while previewing the image. As a result, a black line is drawn around the particles. The Separator filter will sometimes require manual editing and should be used as a guideline.

After the Separator filter has been applied, a threshold must be set to remove background area from the detectable portion of the image. Images collected with photo mode will show two peaks in the histogram. The darker peak should be large and narrow. This corresponds to the distribution of grayscale values in the background of the image. The quantity of background should be high and due to its relatively flat surface, the gray values will be narrow. The toner particles in the image will have higher grey values. Due to the contour changes of the toner particle, the region of interest in the histogram will have a much broader peak ranging from the base of the background peak to a full white value. This is illustrated in Figure 2. The threshold is set from the base of the background peak to a full white value of 255.

The detection algorithm is set to fill holes inside circular regions so that smooth particles that have internal grey values that may interfere with the background range are included in the area. After the detection, manual particle deletion must be done to remove erroneous or overlapped particles. A minimum particle size filter is used to remove any noise that is still remaining after the Median filter. A setting of 300 pixels is effective for this task. At a magnification of 1000X, a 7.0µm particle occupies approximately 3.5e3 pixels. Setting the Detection algorithm to exclude particles that are on the border of the image helps reduce the number of erroneously detected particles.

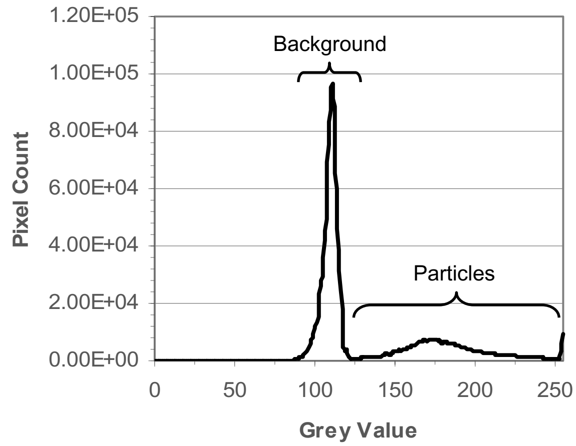

Figure 2. *A histogram of a typical FE-SEM toner image*

During the detection, pixel connectivity must be limited to only adjacent borders since the Separation filter only draws a 1 pixel width line. If the pixel connectivity is set to include diagonals, the Detection algorithm will negate the effects of the Separation filter.

The results specified for use are named "Circularity Index," however they correspond to what is more commonly known in the image analysis field as shape factor. The CI of each particle is determined by the following formula.

$$CI = 4 \cdot \pi \cdot \left(\frac{A_p}{P_p} \right) \tag{1}$$

Where A_p is the area of the particle and P_p is the perimeter. The basis of this formula is determining the difference in the actual particle area and a circle of equivalent area. As the particle becomes less circular, the perimeter will increase or the area will decrease to reflect this change. The results are collected per particle and averaged for the CI value of a particular toner.

Experimental Results

The increase in magnification provided by the SEM provides a more sensitive method for determining the CI of toner particles than previous methods. Figure 3 shows the effect of increasing magnification on the CI of a toner. The local minimum at 500X is attributed to the single pixel width line drawn around the particle by the separator filter. The application of the separation filter at magnifications lower than 500X is impossible due to the relative area of a 1 pixel width line to the area of the particle. Table 1 shows the relationship between magnification and pixel area of a 7.0 µm particle. Table 2 shows that as the circularity index of the particle decreases, the relative area of the line increases. The relative area decrease alone is not significant, but when placed into formula 1, the effect is magnified by further skewing the ratio of the area to the perimeter. In application of the separator filter, some of the gray values that are similar to the background are trimmed from the edges of the particles.

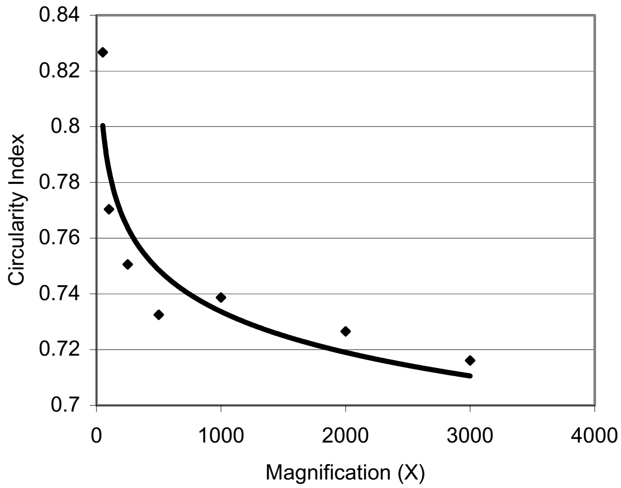

Figure 3. *Circularity as a function of magnification*

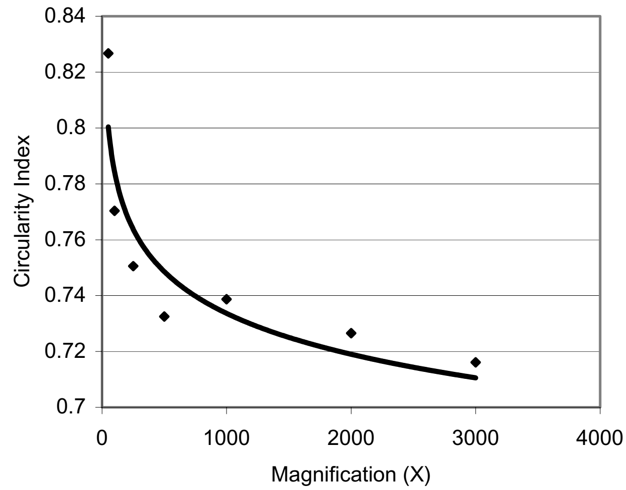

Figure 4. *Circularity values reported by image analysis and flow cytometry*

Table 1: Relative area of a 1 pixel width line at 1280x1024 resolution of a 7.0 µm diameter particle (CI = 1.0)

Magnification (X)	μm^2/ pixel	Pixels/ sphere	Area of line (μm^2)	Area Loss (%)
250	1.76e-1	2.19e2	9.32	4.082
500	4.40e-2	8.76e2	4.62	0.524
1000	1.10e-2	3.50e3	2.31	0.066
2000	2.75e-3	1.40e4	1.15	0.008
3000	1.22e-3	3.15e4	0.77	0.002

Table 2: Relative area of a 1 pixel width line at 1280x1024 resolution of a 7.0 µm diameter particle (CI = 0.75)

Magnification (X)	μm^2/ pixel	Pixels/ sphere	Area of line (μm^2)	Area Loss (%)
250	1.76e-1	2.19e2	13.31	4.841
500	4.40e-2	8.76e2	6.60	0.626
1000	1.10e-2	3.50e3	3.30	0.079
2000	2.75e-3	1.40e4	1.65	0.010
3000	1.22e-3	3.15e4	1.10	0.003

Results from 11 different toner samples were measured with a flow cytometry-based instrument and compared to the shape analysis by FE-SEM imaging. (Figure 4) While the values collected by the flow cytometry-based instrument do correlate to the shape analysis values, the sensitivity and dynamic range are much greater for the image analysis method. The lack of resolution in flow cytometry is illustrated by the artificially inflated circularity values and small range. A toner particle moving through a flow cell with a resolution of 0.7 µm/pixel will occupy 752% less pixels in the CCD camera than the same particle being imaged on an FE-SEM at 1000X. At 1000X, this corresponds to a resolution of 0.09µm/pixel.

By gathering the data per particle, we are also able to create histograms of the particle shapes. This has an advantage over the typical single number average in that the actual particle shape distribution can be seen. This allows toner production methods to be optimized to produce specifically shaped particles and provides the ability to discern between bimodal particle shape distributions. Figure 5 shows cumulative percent histograms of post-processed toner particles. These particles have undergone a heat treatment to increase their circularity index. Each line represents a different configuration in temperature of treatment and feed rate.

Conclusion

FE-SEM image analysis provides a significantly more accurate measurement of particle shape. Because of the heavy influence that particle shape has on powder flow, having an accurate measurement of shape is important to the toner industry. This method can be applied to finished toners to determine their flowability in the toner cartridge just as well as it can be applied to raw materials during manufacturing.

The enhanced resolution provided by the high magnification capabilities of the FE-SEM give this method a much longer lifetime. It is possible to collect images of particles as small as 200 nm, thus providing the ability to handle future toner particle size changes and a method for characterization of toner surface additives.

This method can be easily adapted to determine shape characteristics of larger carrier particles as well by reducing the magnification. As long as the number of pixels per particle is maintained, virtually any size particle that can be imaged with an FE-SEM can be measured.

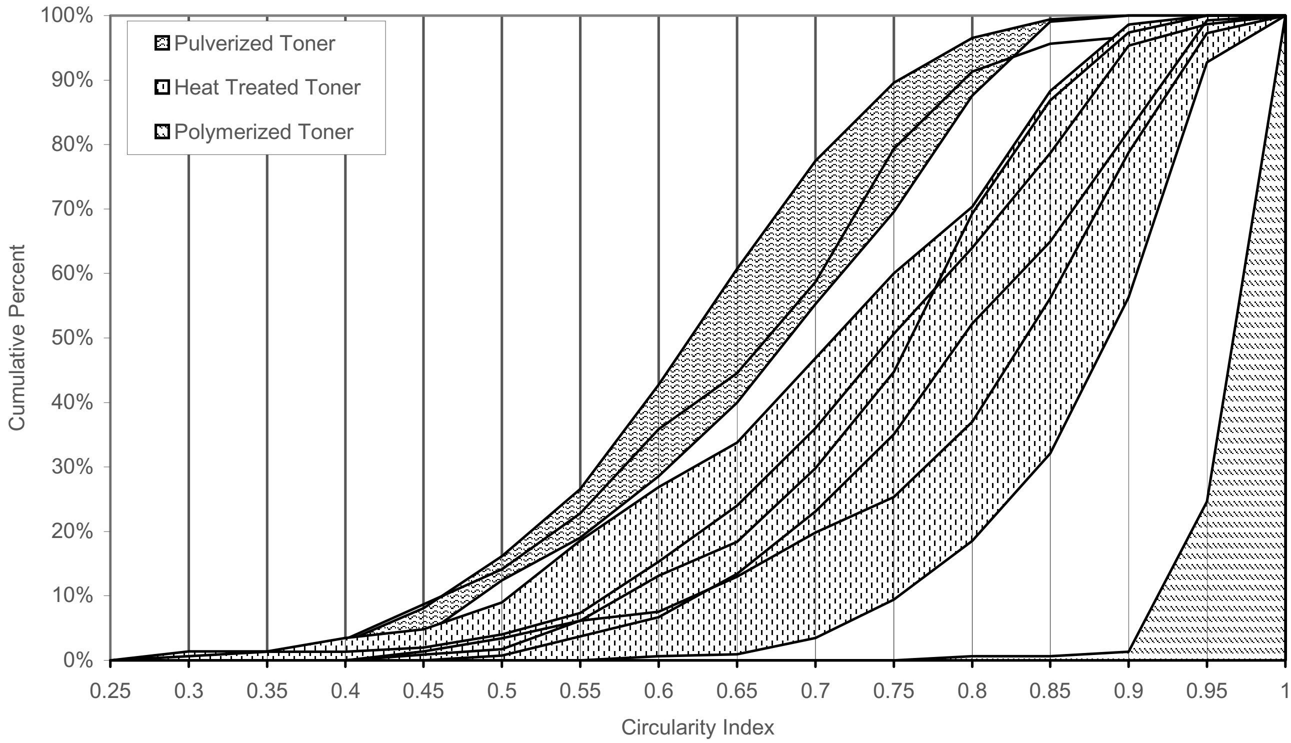

Figure 5. *Effects of post processing for improved CI range.*

References

[1] P.W. Cleary, The Effect of Particle Shape on Hopper Discharge, Second International Conference on CFD in the Minerals and Process Industries, Melbourne, Australia, 1999.

[2] J.C. Russ, The Image Processing Handbook (CRC Press, Boca Raton, FL, 2002).

Author Biography

Dustin Earnhardt joined Mitsubishi Kagaku Imaging Corporation in 2004 as an analytical chemist, researching toner particle shape and surface additive binding energies. Previously, he worked at the National Institute of Standards and Technology on particle transfer and thermal imaging of materials. He received his B.S. degree in Chemistry from Old Dominion University in 2004. Since 2004, he has been a graduate student in the Master's of Materials Science Engineering at the University of Virginia. His primary research interest is analytical microscopy and materials science. He is a member of IS&T.