Comparison of Flour Particle Size Distributions Measured by Electrical Resistivity and Microscopy[1]

J. T. WILSON[2] and D. H. DONELSON, Soft Wheat Quality Laboratory,
Ohio Agricultural Research and Development Center, Wooster, Ohio

ABSTRACT

A comparison was made of procedures for particle size distribution analysis on a group of air-classified flour samples. The procedures were microscopic (with the use of Martin's diameter) and electrical resistivity (with the Coulter counter). Shape correction factors derived by two independent means, using methods of moments, were in good agreement. It appears that the microscopic method is oversizing and a correction factor should be applied to the microscopic data. When this is done, particle size distribution data obtained by the two methods appear to be very similar.

Flour particle size measurements have been obtained by a number of methods including sieving, microscopy, air permeametry, gravitational sedimentation, centrifugal sedimentation, and changes in electrical resistivity. Gracza (1) determined flour and air-classified particle size by several methods and defined a number of particle size distribution measurements. Ames et al. (2) found good agreement between results for a microscopic electronic sizing and counting ("flying spot") method and a liquid sedimentation balance procedure for a group of fine powders. When Irani (3) and Irani and Fong (4) compared data by these two methods, which they considered to be absolute methods, with those by the change in electrical resistivity (Coulter counter) method on flour particles, differences in results were noted. Results were systematically high by the last-mentioned procedure on a weight percent greater than stated diameter basis. The deviations of Coulter counter diameters increased as particle size decreased. Such data for flours could be adjusted by a procedure (3) in which flour results were corrected on the basis of a previously calibrated flour. Measurements between Coulter counter and the absolute methods were in good agreement when glass beads were used. Monocalcium phosphate powder values required adjustments for the Coulter counter values in order to obtain agreement with the other methods. The correction factors for the phosphate powder were different from those calculated for the flour samples.

At this laboratory, the Coulter counter has been used extensively for flour fraction particle size measurements. This paper presents data comparing a microscopic and an electrical resistivity (Coulter counter) procedure for a group of samples. These comparisons were made by two independent approaches.

In one method, linear shape correction factors for the microscopic measurements were calculated; diameter, count, and particle average volume size obtained only from microscopic data were used. In the second method, linear shape correction factors were calculated from various ratios of moments of the uncorrected microscopic and Coulter counter distributions. Shape factors determined by these two methods were thus available for comparison.

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[2]Present address: Department of Biological Sciences, Purdue University, Lafayette, Indiana 47907.

Graphs of particle size distributions for Coulter counter and microscopic measurements are also presented to indicate the effect of shape factor on the microscopic distributions.

MATERIALS AND METHODS

Allis laboratory-milled Seneca soft wheat flour of 50% extraction was air-classified on a laboratory-scale Pillsbury Turbo separator. Particle size by both Coulter counter and microscopic methods were made on the parent flour, the five fine fractions, and the coarse residue for comparative purposes.

Coulter Counter Measurements

Coulter counter measurements were made with the use of a solution of 4% ammonium thiocyanate in anhydrous isopropyl alcohol as suspension medium. A tube with 280-μ aperture was calibrated with corn and ragweed pollens, and one of 140-μ aperture was calibrated with ragweed pollen and 3 μ polystyrene latex The calibrants were measured with a microscope. Shape corrections were made for corn pollen. Coincidence corrections were made by the standard formula which assumes a Poisson distribution for coincidence by number. Blanks for the suspension medium were subtracted from the corrected counts. Adjustments for blanks were important only for first and second fines fractions carrying mass median diameters (MMD) of about 6 and 10 μ, respectively. Intervals usually used were: 2.5-μ intervals to 30-μ size, 5 to 45 μ, and 10 to 95 μ. The actual aperture system used depended on the type of material.

In general, the Coulter counter procedure and the distribution calculations were performed as outlined in the instruction manual (5). The operation of the Coulter counter is also discussed by Berg (6). The effect of coincidence on particle count has also been studied by Princen and Kwolek (7).

Microscopic Measurements

Microscopic measurements were made with a Leitz Ortholux binocular microscope fitted with an eyepiece micrometer calibrated with a ruled-stage micrometer. It was necessary to use a verniered mechanical stage to select randomized fields on the slide. The parameter measured was Martin's diameter (8).

Samples were prepared by mixing flour and isopropyl alcohol on a slide with a wooden toothpick, placing a cover slip over the suspension, and sealing with petroleum jelly. For the first and second fines fractions, about 800 particles were measured and classified by size into 1-μ intervals. About 1,000 particles were sized for the third-, fourth-, and fifth-cut fines because of their larger mean diameters and greater ranges of sizes, and about 1,500 counts for the coarse residue and parent flours, for the same reason. All counts were made in triplicate. For computing frequencies, 2-μ intervals were taken for the first four fines fractions, giving 10 to 20 classes, and for the fifth fine and fifth coarse and parent, 4-μ intervals were taken, giving 12 to 25 classes.

Since flour particles have rather low axial ratios (thickness over length about 0.5), and many are irregular in outline, and since they inevitably orient on a slide with the largest diameter normal to the optical axis, it is necessary to compute shape factors if one wishes results in the form of equivalent spherical diameter. Examples of particle shape factors and methods for determining them are discussed

by Herdan (8) and Orr and DallaValle (9). The standard procedure for this measurement is to make dilutions of a known weight of material and count samples in a haemocytometer counting cell. For this, one needs water or a suspension medium of the proper surface tension to hold the drop between cover slip and ruled ledge. It was found in our work that water did not make a suitable medium with flour fractions which contained 20 and 30% protein. A carbon tetrachloride-chloroform mixture of specific gravity 1.40 gave excellent dispersion and removed the tendency for large particles to settle out rapidly. However, it was found that not only does it evaporate from the counting cell rapidly, but it runs off the raised ledge. A counting cell was finally made from a 100-square ruled eyepiece graticle. After dilutions were made to a concentration of about 500 to 1,500 particles per 0.1 ml., 0.1 ml. of the suspension was pipetted into the net, and after the solvent had evaporated, the particles were counted with incident illumination.

RESULTS

Data obtained by the two methods (microscope and Coulter) were compared by a method of moments. Since intervals selected for tabulating histograms should not matter so long as they are small enough to include variations in the distribution, this is the natural method for comparing the data. There are inevitably differences at any particular size among duplicates and methods, because of sampling error and experimental errors, but if two methods give the same ratios of various combinations of moments they give the same distribution.

There are several interesting results in the comparison of ratios of moments,

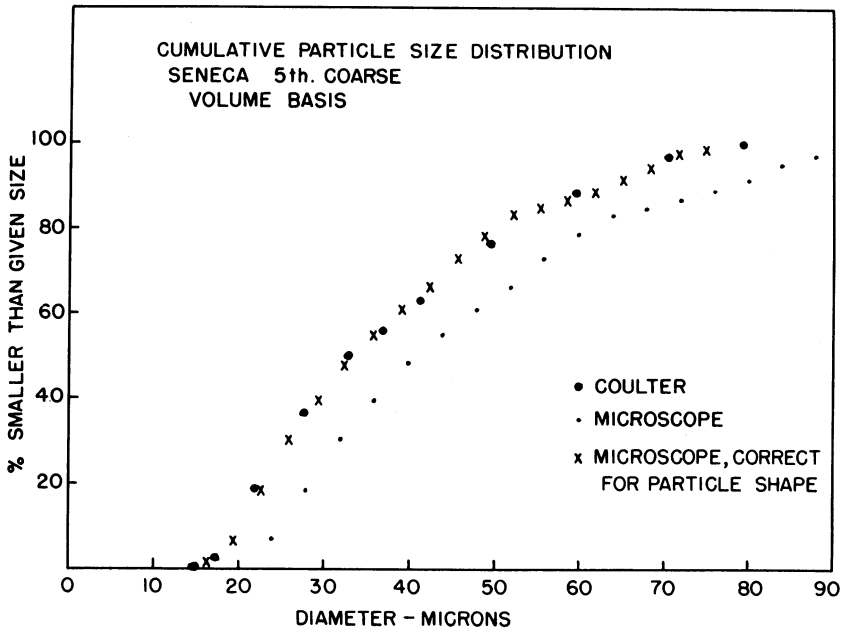

Fig. 1. Comparison between Coulter counter and uncorrected and corrected microscopic particle size distributions for fifth coarse fraction. Distributions on a cumulative weight (volume) basis.

TABLE I. PARTICLE WEIGHT DISTRIBUTIONS FOR THE COULTER COUNTER AND MICROSCOPIC METHODS FOR ONE SET OF FIFTH COARSE FRACTION DATA

Coulter Counter		Microscope		
Diameter μ	Wt. % Finer Than Diam. %	Uncorr. Diameter μ	Corr. Diameter[a] μ	Wt. % finer Than Corr. Diameter %
7.5	0.0	4	3.5	0.0
10.0	0.0	8	7.0	0.0
15.0	0.0	12	10.5	0.1
20.0	9.5	16	14.0	0.4
25.0	25.9	20	17.5	2.3
30.0	40.5	24	21.0	8.4
35.0	52.0	28	24.6	20.5
40.0	63.6	32	28.1	31.0
45.0	70.5	36	31.6	39.5
55.0	86.2	40	35.1	49.0
65.0	95.2	44	38.6	56.1
75.0	98.2	48	42.1	61.9
85.0	99.6	52	45.6	68.6
95.0	99.9	56	49.1	72.3
105.0	100.0	60	52.6	78.1
		64	56.1	83.8
		68	59.6	83.8
		72	63.1	85.9
		76	66.7	88.3
		80	70.2	96.7
		84	73.7	100.0

[a]Corrected diameter, obtained by multiplying uncorrected diameter by shape factor of 0.877.

which are brought out best by first showing some cumulative weight graphs before any mathematical calculations are given. The graphs represent the percent of weight finer (smaller) than the stated diameter. Actually, of course, volume cumulants were calculated from the data.

Figure 1 shows the result of the first pair of measurements with microscopic and electronic methods, during preliminary investigations of the problem of shape corrections. Data are for the fifth coarse where the large dots are the Coulter cumulative weight basis distribution, the small dots are the microscopic distribution uncorrected for particle shape, and the crosses are the corrected microscopic data. The shape factor was 0.87, calculated from microscopic measurements.

An example of one of the sets of numerical values obtained on the fifth coarse fraction by Coulter counter and microscopic measurements is shown in Table I. This table presents the cumulative weight distribution by the Coulter counter method and the corrected microscopic distributions. In this case the microscopic shape factor was 0.877. A list of shape factors and the method for calculating them is presented later.

Figure 2 shows the comparison for distributions between the second fine (10 μ MMD) and fourth fine (18 μ MMD). The same results were obtained by the two methods. It may be noted that the diameter scale on all graphs is linear, and it is not necessary to preshorten the diameter scale with a logarithmic representation to have results by the two methods coincide.

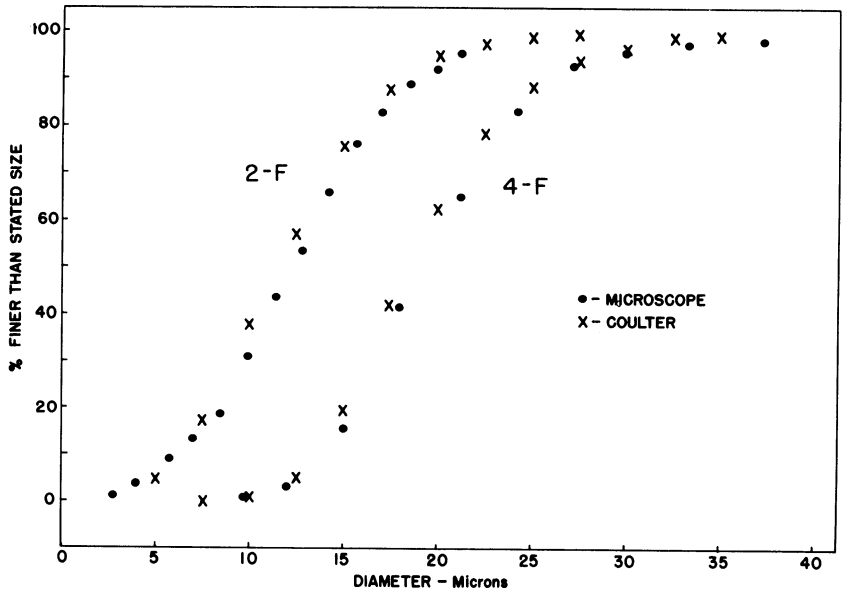

Fig. 2. Comparison between Coulter counter and corrected microscopic particle size distributions for second (2F) and fourth (4F) fines fractions, weight basis.

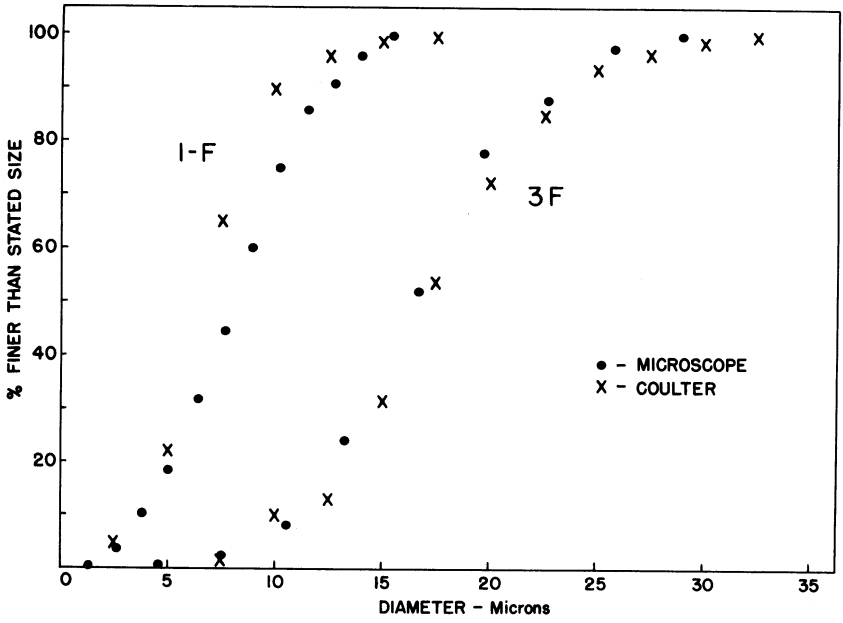

Fig. 3. Comparison between Coulter counter and corrected microscopic particle size distributions for first (1F) and third (3F) cut fines fractions, weight basis.

Figure 3 shows the data for the third fine (MMD about 16 μ) for which very good agreement was obtained between Coulter counter and corrected microscopic data.

Results for the first-cut fines fraction show some deviation, although the comparison is quite good. They indicate also that the Coulter counter results give a greater percentage of particles below 10 μ than the corrected microscopic data. This is just the reverse of the published critique of the electronic method. This is perhaps as it should be. An aperture of 140 μ with interval settings indicated previously was used for these small particles. It is our experience that this is a practical size to use for fines fraction and fine materials in general. For very fine samples such as this first-cut fraction, however, a smaller aperture and another size interval arrangement might have produced somewhat different results.

Figure 4 shows a comparison of the two methods for the Seneca parent flour. Here the correction used has fitted the upper end of the distribution rather well, and improved the rest of the microscopic distribution. However, there is still some deviation below 40 μ, although the hump is present in both the Coulter and microscopic curves.

Probably the fit could be improved by obtaining more data by microscopic sizing. The size range of the flour is 0 to 116 μ, and a considerable number of counts are needed to ensure a representative sample.

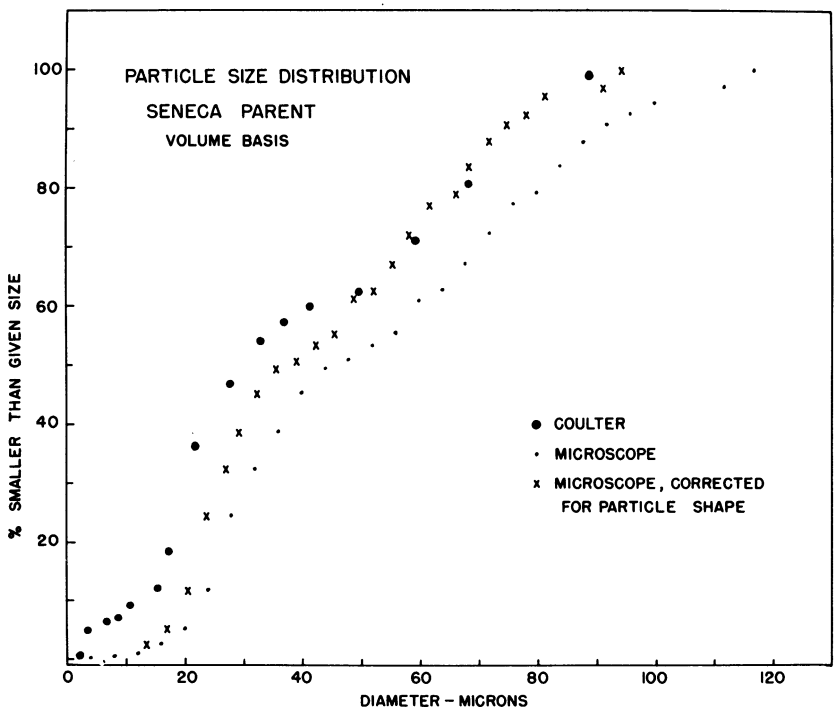

Fig. 4. Comparison between Coulter counter and uncorrected and corrected microscopic particle size distributions for parent flour, weight basis.

TABLE II. ESTIMATION OF PARTICLE SHAPE FACTORS FOR MICROSCOPIC MEASUREMENTS

Flour or Fraction	$(d'_m)^3$ μ^3	ρ g./ml.	N X 10^8/g.	a_v	a_l
Parent	16,268	1.455	2.252	0.358	0.710
First fines	434	1.390	120.6	0.263	0.641
Second fines	1,325	1.431	28.01	0.361	0.712
Third fines	5,114	1.486	5.762	0.438	0.758
Fourth fines	10,967	1.494	2.682	0.436	0.757
Fifth fines	15,432	1.480	1.828	0.456	0.768
Fifth coarse	32,419	1.455	0.5977	0.678	0.877

The values of the shape factors and an example of the calculation involved follow. First, Table II shows the tabulation of numerical values used in computing shape correction factors where

$$a_l = [6/\pi\rho N(d'_m)^3 \times 10^{-12}]^{1/3}$$

gives the linear shape correction. In correcting the moments, one multiplies Σnd terms by a_l, the Σnd^2 terms by a_l^2, the Σnd^3 terms by a_l^3, and the Σnd^4 terms by a_l^4.

An example of the method used for the numerical calculation of shape factors is given below.

$$a_v = 6a'_v/\pi,\ a'_v = 1/\rho N(d'_m)^3,\ (d'_m)^3 = \Sigma nd^3/\Sigma n,\ a_l = (a_v)^{1/3}$$

where a_v = volume shape factor;
ρ = density in g./ml.;
N = number of particles per g. by microscopic count;
d^3 = average of cubed diameters of each end of interval; and
n = number of particles per interval by microscopic count.

For the fifth coarse fraction the calculations were as follows:

$(d'_m)^3$ = 32,419 μ^3,
ρ = 1.455 g./ml.,
N = 5.977 X 10^7 particles per g.,
a'_v = -.3548, and
a_v = 0.6777.

From the value for a_v, a linear shape factor a_l = 0.877.

Now, if we take the ratios of successive moments for the Coulter data and also

TABLE III. RATIOS OF SUCCESSIVE MOMENTS FOR COULTER COUNTER DATA AND FOR UNCORRECTED MICROSCOPIC DATA

Flour or Fraction	d_n		d_l		d_{vs}		d_m	
	Coulter μ	Micro. μ	Coulter μ	Micro. μ	Coulter μ	Micro. μ	Coulter μ	Micro. μ
Parent	15.20	14.79	20.14	27.63	27.65	39.13	38.59	52.74
First fines	4.38	5.14	5.64	7.90	6.76	10.71	7.67	12.65
Second fines	5.86	7.54	8.24	11.54	10.53	14.90	12.55	17.93
Third fines	12.50	12.63	14.44	18.34	16.20	20.99	17.66	23.15
Fourth fines	15.34	19.92	16.70	22.95	18.12	25.13	19.43	27.11
Fifth fines	18.68	19.53	20.10	26.43	21.80	29.50	23.67	31.94
Fifth coarse	24.55	24.83	28.09	33.05	32.93	39.39	38.86	47.10

for the *uncorrected* microscopic data, we obtain various diameters, d_n, d_l, d_{vs}, and d_m, which are defined by the ratios:

$$d_n = \Sigma nd/N, \qquad d_{vs} = \Sigma nd^3/\Sigma nd^2$$
$$d_l = \Sigma nd^2/\Sigma nd \qquad d_m = \Sigma nd^4/\Sigma nd^3$$

The numerical values for these diameters are presented in Table III.

If, further, we take the ratio, for each type of diameter, of the Coulter value for the diameter to the uncorrected ratio for microscopic diameter, d_i (Coulter)/d_i (microscopic), the value should be equal to the shape factor computed for microscopic data.

Table IV shows that this is approximately the case for the ratios of d_{vs} and ratios of d_m values. Both the d_{vs} and d_m diameters involve the third moment of the distributions and the shape correction α_l was computed from the third moment. However, the moments used to calculate the d_n and d_l values are sensitive to sampling variation and counting error. For example, a large variation in particle count at the small size end of a distribution would have only a small effect on the form of a cumulative weight curve, but it could produce considerable change in the cumulative number curve.

TABLE IV. RATIOS OF DISTRIBUTION DIAMETERS, d_i (COULTER)/d_i (MICROSCOPIC), COMPARED TO α_l, THE LINEAR SHAPE FACTOR. THE LINEAR SHAPE FACTOR WAS CALCULATED FROM MICROSCOPIC MEASUREMENTS

Flour or Fraction	Distribution Diameter Ratio				
	d_n	d_l	d_{vs}	d_m	α_l
Parent	1.03	0.73	0.71	0.73	0.71
First fines	0.85	0.71	0.63	0.61	0.64
Second fines	0.78	0.71	0.71	0.70	0.71
Third fines	0.99	0.79	0.77	0.76	0.76
Fourth fines	0.77	0.73	0.72	0.72	0.76
Fifth fines	0.96	0.76	0.74	0.74	0.77
Fifth coarse	0.99	0.85	0.84	0.83	0.88

DISCUSSION

The linear shape correction factors, derived through two independent observational and computational procedures, have thus been found to be very closely related, indicating that they represent values approximating acceptable magnitudes. From a consideration of parameters obtained by the Coulter counter (change in electrical resistivity dependent on volume displacement of medium by particles) and by the microscope (measurement of Martin's diameter of particles), it appears likely that the latter method is oversizing, and that the correction factors should therefore be applied to microscopic data. When this is done, particle size distribution data obtained by the two methods appear to be in good agreement.

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