

A comparative study of particle size analysis in fine powder: The effect of a polycomponent particulate system

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Abstract—We report the particle size distribution of poly component particulate systems studied in three kinds of experimental methods. Six analyzers, such as Mastersizer Microplus (Malvern Instruments Ltd., UK), LS230 (Coulter Electronics Ltd., USA) LMS30 (Seishin, Japan), Analysette22 (Fritsch, Germany), HELOS (Sympatec, Germany) based on a laser diffraction and scattering method, and the SKC-2000S (Seishin Co., Ltd., Japan) based on the centrifugal sedimentation method, were used to study the particle size distribution. The results do not show reasonably good agreement between the different analyzers and different sample systems. There is a discrepancy regarding the absolute values, which can be explained by the fact the techniques used are based on different measuring principles. The results of the present study reflect that the investigator must carefully select the particle size analyzer for a particular application. Therefore, we suggest that it is necessary to measure the particle size distribution by using at least two types of analyzers or more of different makers.

Key words: Laser Scattering and Diffraction, Particle Size Analysis, Polycomponent Particulate System

INTRODUCTION

The particle size distribution (PSD) of particulate materials is an important parameter that governs the proper function of many industrial products such as metal powders, drug formulations, lattices, pigments, and ceramic materials. From the perspective of obtaining a better handle on manufacturing processes, it is essential to define the characteristics of particulate materials in a particular state in order to draw up specifications and quality levels which can be acceptable for raw materials [1-4]. Many kinds of instruments have been developed and used in various industrial fields [5,6] to measure the particle size distribution in order to achieve better manufacturing.

In the last few years, the measurement techniques of particle size distribution of fine particles have also progressed together with the development of laser technology and hardware and software for personal computers [7]. Many kinds of instruments for particle size measurement based on physical principles such as visual observation, Stokes's sedimentation law, laser diffraction and scattering theory and electrical counting of particles have been developed and used. From the research point of view, it is interesting to do a comparative study of the particle size distribution of a particulate system by using different types of particle size analyzers.

We have investigated the particle size distribution of a polydisperse particulate system and mixed samples of two or three mono dispersion particles. Particularly, the aim of the present study was to see the influence on the particle size distribution using different apparatus. For this reason, we have used a different apparatus for

each test sample [8-14].

EXPERIMENTS

The samples used in the first study were two kinds of calcite powders. One consisted of fine particles (FP1) of $x_{50}=0.42-0.62\ \mu\text{m}$ in size and another consisted of coarse particles (CP1) of $x_{50}=47.80-60.39\ \mu\text{m}$ in size. The calcite powders of two monocomponent particles with significantly different median diameters, 1 : 1, 1 : 2 and 2 : 1, based on mass, were examined and compared with the calculation value of particle size distribution and experimental ones. To compare the particle size distribution of these samples, we measured and analyzed the particles using different analyzers such as the Mastersizer Microplus (manufactured by Malvern Instruments Ltd., UK), LS230 (manufactured Coulter Electronics Ltd., USA) based on a laser diffraction and scattering method, SKC-2000S (Seishin Co., Ltd., Japan) based on the centrifugal sedimentation method.

In the second experiment, an analysis was made using the fine particles (FP2, AMS-9) in the submicron range ($x_{50}=0.47-0.85\ \mu\text{m}$), middle particles (MP, AM-21) of $x_{50}=3.20-5.89\ \mu\text{m}$ and coarse particles (CP2, CF-50F) of $x_{50}=54.48-63.89\ \mu\text{m}$. In the sample mixing method of the second study, we selected four kinds of poly-component particulate systems, 1 : 1 : 1, 1 : 1 : 2, 1 : 2 : 1 and 2 : 1 : 1, based on mass. The particle size distribution of these systems was measured, analyzed and compared by using the following setup: Mastersizer Microplus (Malvern Instruments Ltd., UK), LS230 (Coulter Electronics Ltd., USA), Analysette22 (Fritsch, Germany), LMS30 (Seishin, Japan), and HELOS (Sympatec, Germany) based on a laser diffraction and scattering method.

In the third experiment, we studied two kinds of calcite powders

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as test samples. This included the ground calcite S500 ($x_{50}=10.26-13.26\ \mu\text{m}$) made by Wangpyo Chemical Co., Ltd., Korea, and precipitated calcite silver-white ($x_{50}=1.65-3.08\ \mu\text{m}$) made by Shiraishi Kogyo Co., Ltd., Japan. The sample mixing methods of the third study were 1 : 1, 1 : 3, 1 : 9, 3 : 1 and 9 : 1, based on mass. The particle size distribution of these systems was measured, analyzed and compared by using the following setup: Mastersizer Microplus (Malvern Instruments Ltd., UK), the LS230 (Coulter Electronics Ltd.,

USA) and the Analysette22 (Fritsch, Germany) based on a laser diffraction and scattering method.

RESULTS AND DISCUSSION

Tables 1-3 show the comparison of the results of particle size, x_{10} , x_{50} and x_{90} , of particle size distribution with various methods and a particle size analyzer for the monocomponent samples and mixed samples. The measurement results of the monodispersed samples are similar; however, the mixed samples are not in good agreement. These results may be attributed to the configuration or the software of the equipment. This is a significant problem in particle size measurement. None of the methods tested in the current work provided a reliable value on the median diameter of particle size distribution measurement. This means that the scatter of data measured should be discussed in detail.

1. First Experiment

Fig. 1 shows a typical particle size distribution of each sample measured with various particle size analyzers. A direct qualitative comparison of the particle size distribution curves shows that Mastersizer, LS230 and SKC-2000S give similar results in all cases, but that microscopy with image analysis and laser methods cannot be considered similar on a mass basis. Particle size distribution on a mass basis is proportioned into three powers of particle size on a number basis. Accordingly, the frequency of large particles is measured at a high percentage. However, it is not easy to compare the three techniques' results of particle size analysis on a mass basis because the laser diffraction, scattering methods and microscopy method have very different measuring principles. In a polycomponent particulate system, samples of fine and coarse particles, fine

Table 1. Comparison of the results of particle size, x_{10} , x_{50} and x_{90} , of PSD using various methods and particle size analyzer for monocomponent samples and mixed samples in first experimental method [unit= μm]

Analyzer		Mastersizer	LS230	SKC-2000S
FP1	x_{10}	0.42	0.17	0.22
	x_{50}	0.62	0.42	0.69
	x_{90}	0.88	5.98	2.84
CP1	x_{10}	40.79	45.17	32.26
	x_{50}	57.75	60.39	47.80
	x_{90}	77.21	80.16	64.60
FP1 : CP1 1 : 1	x_{10}	0.49	0.21	0.46
	x_{50}	30.25	48.88	29.01
	x_{90}	78.54	80.35	46.04
FP1 : CP1 1 : 1	x_{10}	0.53	0.19	0.49
	x_{50}	44.54	42.15	44.37
	x_{90}	82.65	69.99	48.42
FP1 : CP1 1 : 1	x_{10}	0.48	0.21	0.50
	x_{50}	1.39	51.38	12.91
	x_{90}	71.44	80.16	45.27

Table 2. Comparison of the results of particle size, x_{10} , x_{50} and x_{90} , of PSD using various methods and particle size analyzer for monocomponent samples and mixed samples in second experimental method [unit= μm]

Analyzer		Mastersizer	LS230	LMS30	Helos	Anaysette 22
FP2	x_{10}	0.18	0.20	0.21	0.31	0.41
	x_{50}	0.47	0.51	0.67	0.64	0.85
	x_{90}	1.32	1.49	1.92	1.24	2.42
MP	x_{10}	0.42	1.72	1.59	0.24	1.11
	x_{50}	5.89	4.69	4.02	3.20	4.45
	x_{90}	15.54	17.65	8.76	7.35	19.81
CP2	x_{10}	48.28	45.05	38.50	42.30	6.66
	x_{50}	63.89	63.85	56.08	59.46	54.48
	x_{90}	86.19	83.43	79.60	83.60	81.77
FP2 : MP : CP2 1 : 1 : 1	x_{10}	0.40	0.28	0.23	0.43	0.92
	x_{50}	54.33	3.96	0.96	2.66	7.49
	x_{90}	98.89	50.26	12.76	14.63	65.90
FP2 : MP : CP2 1 : 1 : 2	x_{10}	0.17	0.39	0.23	0.39	1.13
	x_{50}	25.45	11.23	0.88	1.18	15.63
	x_{90}	79.02	75.41	9.21	7.19	69.69
FP2 : MP : CP2 1 : 2 : 1	x_{10}	0.17	0.30	0.23	0.38	0.94
	x_{50}	4.38	3.28	1.00	1.36	5.58
	x_{90}	57.46	56.60	54.52	3.59	60.11
FP2 : MP : CP2 2 : 1 : 1	x_{10}	0.15	0.26	0.23	0.33	0.78
	x_{50}	1.08	1.27	0.93	0.75	5.55
	x_{90}	59.79	16.25	48.90	1.61	62.58

Table 3. Comparison of the results of particle size, x_{10} , x_{50} and x_{90} , of PSD using various methods and particle size analyzer for monocomponent samples and mixed samples on third experimental method [unit= μm]

Analyzer		Mastersizer	LS230	Analysette-22
S500	x_{10}	0.45	1.21	1.26
	x_{50}	10.26	11.38	13.26
	x_{90}	23.19	25.10	25.40
S-W	x_{10}	0.30	1.52	0.80
	x_{50}	1.65	3.08	2.51
	x_{90}	5.51	5.45	4.78
S500 : S-W 1 : 1	x_{10}	0.31	1.41	0.86
	x_{50}	2.53	3.75	3.12
	x_{90}	15.72	17.95	15.36
S500 : S-W 1 : 3	x_{10}	0.30	1.40	0.73
	x_{50}	1.84	3.29	2.13
	x_{90}	7.18	8.51	3.64
S500 : S-W 1 : 9	x_{10}	0.29	1.39	0.81
	x_{50}	1.69	3.06	2.55
	x_{90}	5.81	5.45	4.99
S500 : S-W 3 : 1	x_{10}	0.33	1.37	1.01
	x_{50}	3.38	5.20	5.20
	x_{90}	21.34	21.52	23.43
S500 : S-W 9 : 1	x_{10}	0.36	1.38	1.06
	x_{50}	6.44	7.95	8.73
	x_{90}	24.30	23.00	24.30

particles are not expressed by the microscopy measurement on a mass basis.

The calculated cumulative particle size distribution of mixed samples for the first experiment was obtained from the following equation:

$$Q_3(x) = \Phi_{FP1} Q_{3,FP1}(x) + (1 - \Phi_{FP1}) Q_{3,CP1}(x), \quad (1)$$

where Φ_{FP1} is mass fraction of the fine powder for the mixed sample, and $Q_{3,FP1}$ and $Q_{3,CP1}$ are the cumulative distribution of each raw sample, respectively [15].

The degree of agreement between the calculated particle size distribution and the experimental ones worsened when utilizing the Mastersizer, LS230, SKC-2000S, and microscope in the ascending order. A direct qualitative comparison of the distribution curves shows that Mastersizer, LS230 and SKC-2000S give similar results in all cases. However, microscopy with image analysis is considerably different due to the difficulty in preparing a slide glass sample having uniform mixing number concentration for optical microscopy.

2. Second Experiment

In the case of monocomponent samples, the measuring curves show very similar results for each piece of equipment. However, in the case of mixed samples, the results of particle size do not really accord with the preparation ratio of the samples, which may be due to the following reasons:

(1) The particle dispersion in the fine particle area occurred with certain particle size distribution which was not measured directly. However, in this case, the monocomponent particulate system and polycrystalline particulate system were repeated.

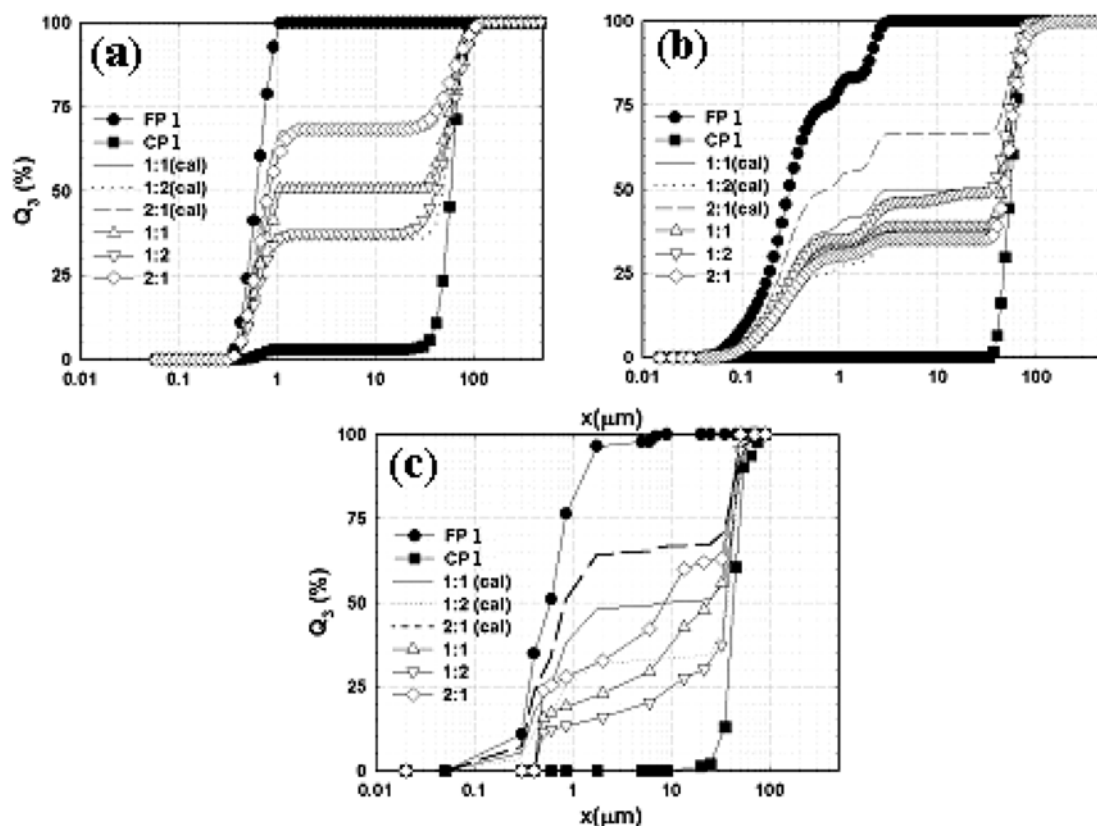


Fig. 1. Comparison of cumulative particle size distribution of 1st experimental using different particle size analyzers.
(a) mastersizer, (b) LS230, (c) SKC-2000S

(2) When the particle was circulated in the particle size analyzer, coarse particle circulation was difficult. In this case, many of the fine particles in the fine particle area were measured more often than coarse particles.

(3) In the case of the dry method of the particle size analysis, the coarse particles and middle particles may have been worn by the circulating nozzle of the particle size analyzer. In this case, the ratio of fine particles was increased. Therefore, in the dry method, one must consider the relationship between the particle size analyzer and

particle characteristics more carefully.

Fig. 2 also shows a typical example of cumulative particle size distribution of mono ingredients and of all components with the value of calculation and measurement for each piece of equipment.

Here, the calculated cumulative particle size distribution of mixed samples for the second experiment was obtained from the following equation:

$$Q_3(x) = \Phi_{FP2} Q_{3,FP2}(x) + \Phi_{MP} Q_{3,MP}(x) + \Phi_{CP2} Q_{3,CP2}(x) \quad (2)$$

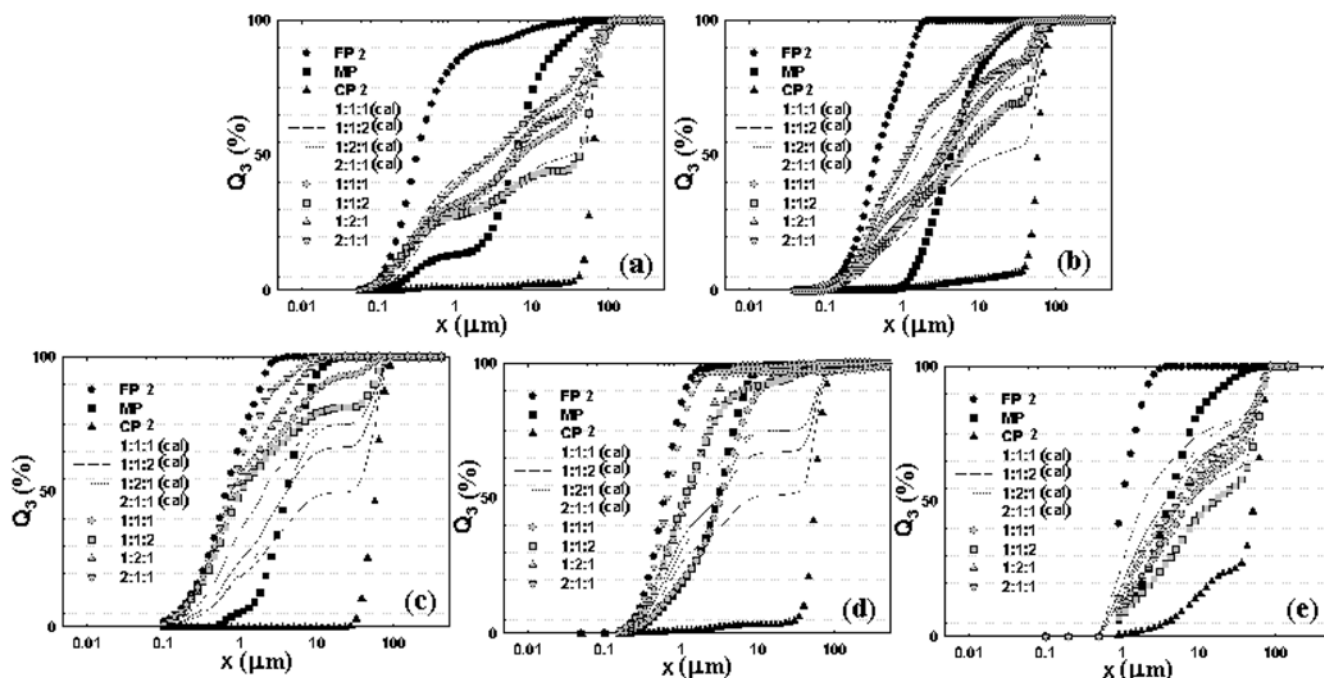


Fig. 2. Comparison of cumulative particle size distribution of mixed samples in second experimental using different particle size analyzers. (a) mastersizer, (b) LS230, (c) LMS30, (d) Analysette-22, (e) HELOS

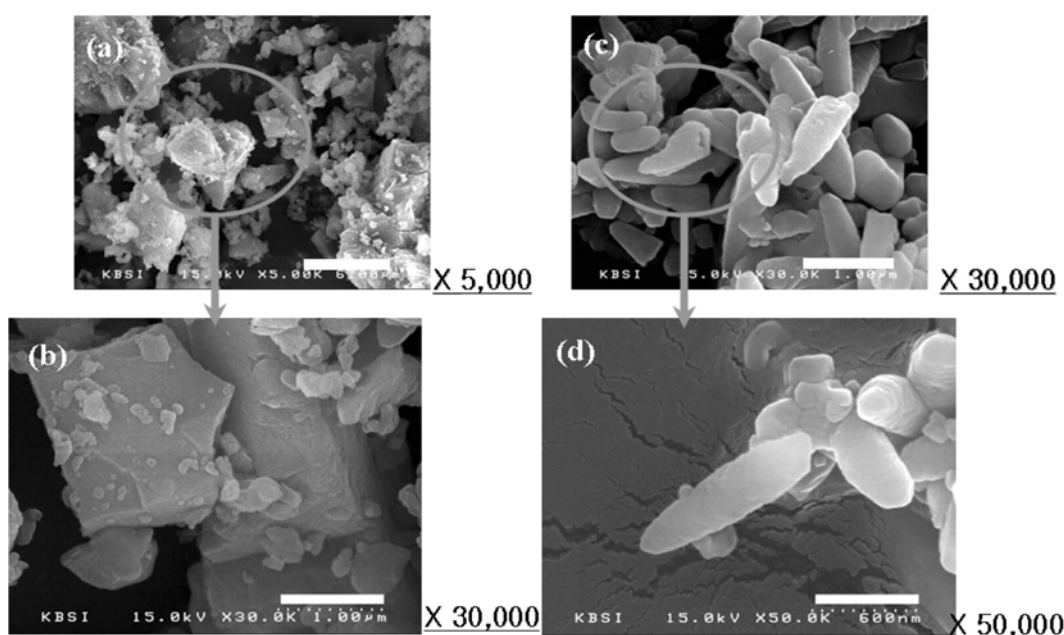


Fig. 3. Typical micrograph of third experimental samples (a), (b) S500 and (c), (d) S-W.

$$\Phi_{FP2} + \Phi_{MP} + \Phi_{CP2} = 1, \quad (3)$$

where Φ_{FP2} , Φ_{MP} and Φ_{CP2} are mass fraction of the mono component powder for the mixed sample, and $Q_{3,FP}$ and $Q_{3,CP}$ are the cumulative distribution of the single component sample, respectively.

The comparison of the calculated and measurement values revealed some differences. The difference between the calculated and experimental value for each piece of equipment did not appear to be so great; however, the cumulative particle size distribution of the monocomponent almost showed the same value without large deviation. From this, it can be concluded that one can measure the particle size distribution of particles of narrow shape ranges. The measured value of the particle size distribution is more or less similar for all of the particle size analyzers.

3. Third Experiment

Fig. 3 shows the typical SEM photographs of two types of calcite samples used to investigate the effects of particle shapes in the present study. Ground calcite S500 was made by Wang Pyo Chemical Co., Ltd., Korea, and precipitated calcite silver-white (S-W) was made by Shiraiishi Kogyo Co., Ltd., Japan. It can be clearly seen from the micrograph that the particle shape of calcite S500 with x_{50} of 10.0 μm is irregular and blocky and its ratio of longest length to shortest length is less than 2.0. However, in case of calcite S-W with x_{50} of 2.0 μm , the shape of the particle is like a spindle, and its ratio of length to diameter of the particle is more than 2.0.

Fig. 4 also shows the comparison of cumulative particle size distribution of raw samples together with three mixed samples. The calculated cumulative particle size distribution of three kinds of mixed samples for the third experiment was obtained from the following equation:

$$Q_3(x) = \Phi_{S500} Q_{3,S500}(x) + \Phi_{SW} Q_{3,SW}(x) \quad (4)$$

$$\Phi_{S500} + \Phi_{SW} = 1, \quad (5)$$

where Φ_{S500} and Φ_{SW} are mass fraction of the calcite S500 and calcite SW for three mixed samples, and $Q_{3,S500}$ and $Q_{3,SW}$ are the cumulative distribution of each monocomponent sample, respectively. As the mass fraction of spindle-shaped calcite SW is increased from 10% to 90%, it is found that there is a deviation in experimentally observed and calculated data of particle size distribution. These results demonstrate that the simple linear additive rule may not apply when expressing the cumulative particle size distribution of samples

including irregular particle shapes far away from spherical particle shapes.

CONCLUSION

We have investigated the particle size distribution of a polydisperse particulate system and mixed samples of two or three monodispersion particles. Particularly, the aim of the present study was to see the influence on the particle size distribution by using different apparatus. For this reason, we have used a different apparatus for each test sample. The comparative study of methods to determine particle size distribution for poly component particulate systems using different analyzer suggests that without great attention, the result obtained from any given instrument may not describe the particulate system for a certain purpose and usage with accuracy. The results of this study also suggest that investigators must carefully select the appropriate particle size analyzers for given applications. It is strongly recommended that the observation of particle size distribution should be conducted in two steps at least: the objective of the first observation step is to identify the total range of the size distribution itself, while the objective of the second step is to examine the size composition of both fine and coarse range in more detail and for more precision, depending on the needs of the investigation and the possible existence of particles in the respective domains.

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NOMENCLATURE

- q_3 : frequency percent under particle size base on volume [%/ μm]
- Q_3 : cumulative percent under particle size base on volume [%]
- x : particle diameter [μm]
- x_{10} : 10% particle diameter based on weight [μm]
- x_{50} : 50% particle diameter based on weight (=median diameter) [μm]
- x_{90} : 90% particle diameter based on weight [μm]
- Φ_x : mass fraction of sample x for mixed sample [-]

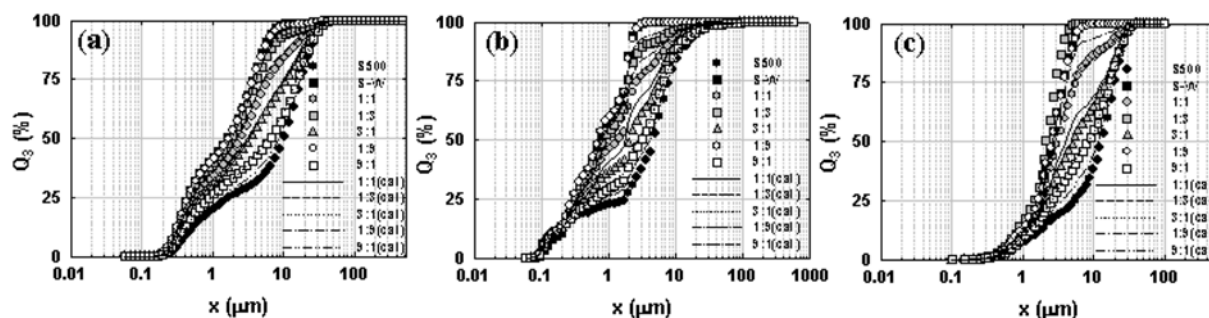


Fig. 4. Comparison of cumulative particle size distribution of the mixed samples of third experimental using different particle size analyzers.

(a) mastersizer, (b) LS230 and (c) Analysette-22

REFERENCES

1. T. Allen, *Particle size measurement*, 4th ed. Chapman & Hall, New York, p.1 (1990).
2. L. Heinz, *Part. Part. Syst. Charact.*, **12**, 148 (1995).
3. C. Andres, P. Reginault, M. H. Rochat, B. Chaillot and Y. Pourcelet, *Int. J. Pharm.*, **144**, 141 (1996).
4. F. Muller, F. Reinhard and T. Polk, *Powder Technology*, **105**, 2 (1999).
5. H. Lange, *Part. Part. Syst. Charact.*, **12**, 148 (1995).
6. F. M. Etzler and M. S. Sanderson, *Part. Part. Syst. Charact.*, **12**, 217 (1995).
7. H. Saveyn, T. L. Thu, P. Govoreanu and P. Van der Meeren, *Part. Part. Syst. Charact.*, **23**, 145 (2006).
8. W. S. Choi, H. Y. Chung, S. S. Kim, B. S. Heo and M. R. Lee, *Pusan Bull. Pharm. Sci.*, **31**(2), 51 (1997).
9. W. S. Choi, H. Y. Chung, B. R. Yoon, J. H. Kang, S. S. Kim and H. S. Kim, *Pusan Bull. Pharm. Sci.*, **33**, 31 (1999).
10. H. K. Choi, S. S. Kim, E. O. Kwak, J. S. Kwak and W. S. Choi, *Pusan Bull. Pharm. Sci.*, **35**, 17 (2001).
11. J. H. Choi, S. J. Ha, Y. C. Bak and Y. O. Park, *Korean J. Chem. Eng.*, **19**, 1085 (2002).
12. H. K. Choi, J. S. Ha and W. S. Choi, *Pusan Bull. Pharm. Sci.*, **37**, 1 (2003).
13. H. K. Choi, J. S. Ha and W. S. Choi, *Pusan Bull. Pharm. Sci.*, **38**, 1 (2004).
14. M. J. Park and J. D. III. Francis, *Korean J. Chem. Eng.*, **21**, 168 (2004).
15. Japan Powder Technology association, *Particle size analysis and technology*, 1st Edition, pp. 150 (1994).