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MICROENCAPSULATED CLOUD SEEDING MATERIALS

Jerrold L. Anderson

Research and Development
Capsular Products Division
The National Cash Register Company
Dayton, Ohio 45409

Contract No. F-19628-70-C-0011

Project No. ILIR, 8620

Task No. ILIR7-69, 862008

Work Unit No. ILIR0001, 86200801

FINAL REPORT

Period Covered: August 15, 1969 through January 14, 1971

February, 1971

Contract Monitor: Loren D. Nelson
Meteorology Laboratory

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This research was supported by the AFCRL Laboratory Director's Fund.

Prepared for

Air Force Cambridge Research Laboratories
Air Force Systems Command
United States Air Force
Bedford, Massachusetts 01730

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ABSTRACT

A process was developed for encapsulating hygroscopic, cloud-seeding agents, sodium chloride and urea. The encapsulation process involves depositing the coating polymer, ethylcellulose, onto finely divided powders using a phase separation-coacervation technique. The process produces small encapsulated aggregates, the size and distribution of which can be varied. Encapsulated materials prepared by the process are unique in that they are void, essentially, of powder fines. Laboratory tests of both encapsulated sodium chloride and urea proved the powders to be resistant to clumping and caking associated with premature moisture sorption occurring during storage and handling. The encapsulation coating polymer, ethylcellulose, is water insoluble, but is permeable to water vapor, water and solutes. Hence, water sorption and particle growth properties resulting from exposure of the encapsulated particles to humid atmospheres are effected by diffusion-mass transport processes. Laboratory testing of encapsulated sodium chloride and urea materials in simulated warm fog atmospheres revealed water sorption characteristics which were comparable to unencapsulated particles having equivalent dimension.

Twenty thousand pounds of encapsulated urea, nominally, 20-80 microns in diameter, were produced and delivered for warm fog dissipation testing conducted by the Air Force Cambridge Research Laboratories. Results of the field tests show that the encapsulated urea functioned effectively and as designed.

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FOREWARD

This report was prepared by Research and Development, Capsular Products Division, The National Cash Register Company, Dayton, Ohio, under Contract No. F19628-70-C-0011. The work was administered under the direction of Mr. Loren D. Nelson, Air Force Cambridge Research Laboratories, Hanscom Field, Bedford, Massachusetts.

The report covers the work conducted from August 15, 1969, to January 14, 1971. The program manager was Mr. Jerrold L. Anderson, Project Leader, Research and Development, Capsular Products Division, The National Cash Register Company.

The National Cash Register Company wishes to thank the personnel of Air Force Cambridge Research Laboratories, Meteorology Research, Inc., and McClellan Air Force Base who participated in the fog dissipation field testing program. Especially appreciated were the suggestions, assistance and efforts extended during the program by Messrs. L. D. Nelson and B. A. Silverman. Air Force Cambridge Research Laboratoties, and Messrs. W. Wilson, W. Carlson, Dr. T. Smith and Dr. A. Weinstein of Meteorology Research, Inc.

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SECTION I

INTRODUCTION

In July, 1969, The National Cash Register Company (NCR) was awarded a contract by the Electronic Systems Division of the Air Force Systems Command for the research and development of microencapsulated cloud seeding materials. The objective of the R&D program was to develop microencapsulation techniques for hygroscopic fog seeding materials to achieve:

(1) particle size control.

(2) increased stability to clumping and caking during storage and handling of the hygroscopic particles.

(3) Effective water sorption by the particles upon exposure to humid warm fog environments.

The program was to include, but not be limited to, sodium chloride, and the preferred particle size range was 20-80 microns diameter. Completion of the program originally called for the production of 6,000 pounds of select, encapsulated material for field tests to be conducted by the Air Force Cambridge Research Laboratories (AFCRL). The test quantity requirement subsequently was extended during the program to 20,000 pounds of encapsulated material. The material of choice was to be selected by AFCRL on the basis of the R&D results in conjunction with the results of AFCRL's numerical model studies of fog dissipating capabilities.

The technical approach taken by Research and Development, Capsular Products Division, was to utilize NCR microencapsulation processes involving non-aqueous, phase separation-coacervation phenomena. Although some of these processes are employed by NCR in the manufacture of various encapsulated products, the aforementioned objectives for cloud seeding materials presented new and unique problems requiring R&D efforts. For instance, microencapsulation of this general type (non-aqueous, phase separation-coacervation) had not been utilized explicitly for particle size control. Stabilization of materials to moisture is a particular property that had been readily accomplished with encapsulation, and also, the flow properties of powders had been improved by encapsulation. However, the problem presented for hygroscopic fog-seeding application was unique in that somewhat contradictory properties were required. Resistance to the effects of moisture was needed to insure against caking and clumping during storage and handling followed by rather uninhibited water sorption of the particles when dispersed into a cloud.

Other R&D considerations involved the development of laboratory test methods for evaluating the efficacy of the encapsulated materials. Additional influencing factors receiving consideration included economy aspects of any newly developed encapsulation process, existing pilot- and production-plant equipment and their operating schedules.

Following is described the technical activities which were conducted in view of the above considerations, and others, for the development of microencapsulated cloud seeding materials.

SECTION II

TECHNICAL DISCUSSION

A. PARTICLE SIZE FEASIBILITY STUDIES

1. General Considerations

The immediate concern relative to the program objectives was the development of encapsulation systems capable of producing the desired particle size range. Based on prior encapsulation experience, two facts were of high importance regarding the initial experimentation. Those facts were:

(1) Existing encapsulation systems of the phase separation-coacervation type were generally aggregation prone. That is, the encapsulation of small particles generally yielded encapsulated aggregates greater in size than the fundamental particles of the starting material. Consequently, the effective, economical encapsulation of particles less than 100 microns in diameter was problematic.

(2) The particle size of any encapsulated product was highly dependent upon the size and size spectrum of the material prior to encapsulation.

Relative to the aggregation problem, two encapsulation coatings and/or processes had shown some promise in previous work for accomplishing effective small particle encapsulation. The coating polymer, ethylcellulose, was one candidate and is a water-insoluble, water-permeable cellulose ether. The water sorption properties of the encapsulated, hygroscopic particles would, therefore, be diffusion-based and would be, among other things, time dependent.

The second coating polymer, cellulose acetate hydrogen phthalate (CAP), is a cellulose ester having acid functionality. The polymer is permeable to water, insoluble in neutral or acidic aqueous media. Formation of the salt form of the polymer, however, can convert the polymer to the water-soluble form. Water sorption properties of encapsulated hygroscopic particles, then, could be dependent upon the acidity or basity of the hygroscopic material. Sorption would be diffusion-based and comparable, in mechanism, to ethylcellulose for neutral or acidic solutions of hygroscopic materials. In alkali solutions, however, the polymer swells readily and eventually can dissolve. Water sorption of hygroscopic alkaline salts might be quasi-instantaneous should the polymer swell (and, therefore, become more permeable) or dissolve in the solution formed upon deliquesence of the encapsulated particle.

Regarding the particle size of the hygroscopic material to be encapsulated, the feasibility of grinding the material to the appropriate size was investigated. A practical method, it was believed, might include wet-milling the material in the encapsulation solvent media, thereby eliminating any need of drying the product prior to encapsulation. Another method considered for achieving the desired particle size was that of encapsulating aqueous solutions of the hygroscopic material. Particle size control would be achieved, therefore, by emulsification techniques rather than by grinding or milling. Subsequent to encapsulation of the aqueous solutions would be the dehydration of the resultant particles to yield maximum water sorption capacity for the fog-seeding agent.

2. Encapsulation of Milled Particles

(a) Ethylcellulose-Coated Sodium Chloride

It was found in preliminary investigations that small particle encapsulations could be prepared by employing a particular polymer-solvent combination. It was found that ethylcellulose polymers having a low degree of substitution (low ethoxyl content) were insoluble in perchloroethylene at room temperature and soluble at elevated temperatures. This theta solvent behavior at reasonable temperatures was attractive in that it provided a simple, effective means of effecting polymer phase separation (and encapsulation). Also, the solvent had the added attraction of being economical and did not present particular processing hazards. Continuing further, it was found that relatively small particles could be encapsulated by: dispersing the particles in a mixture of ethylcellulose and perchloroethylene at room temperature; heating the mixture to 80-90°C to dissolve the polymer; cooling the system to room temperature thereby effecting phase separation of the polymer and encapsulation. The encapsulated particles were collected by filtration, and the residual solvent was removed by evaporation.

In practice, small particle sodium chloride encapsulations were prepared by milling (using a laboratory attritor) crystalline particles in a portion of the perchloroethylene solvent; by washing the milled particles into an agitator-equipped encapsulation vessel with additional solvent; by adding ethylcellulose; and by conducting the heating and cooling cycles.

The particle size distribution of a representative batch, CRL-A23, as determined with Sharples Micromerograph is shown in Figure 1. The median diameter is observed to be 18 microns. Plotting the weight-size data on a log probability scale and calculating the statistical mean volume diameter, d_V, the value becomes 15.0 microns, with a standard deviation, σ, of 8.1 microns. These initial results established that small particle encapsulations were feasible using finely divided sodium chloride particles and the coating polymer, ethylcellulose.

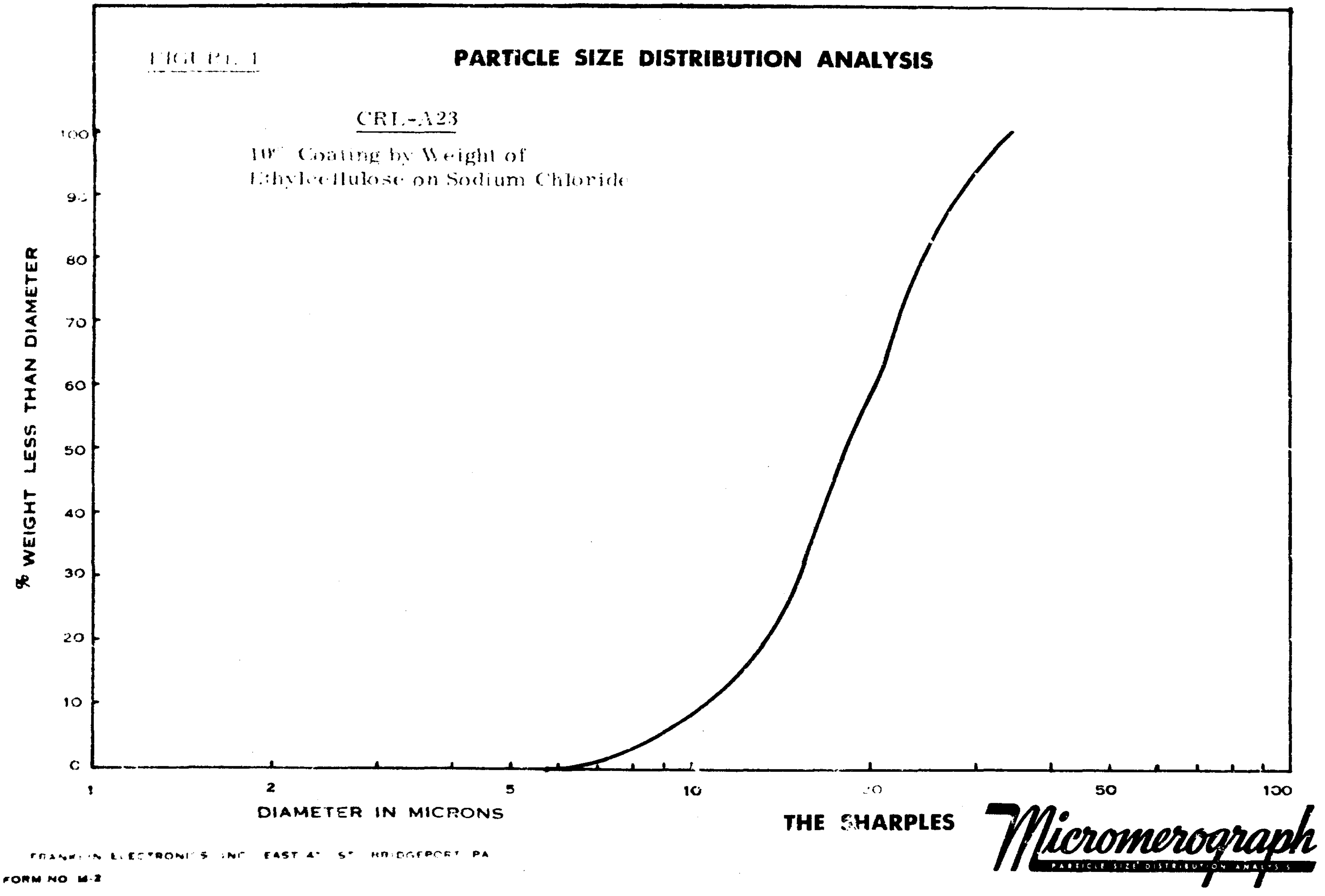

FIGURE 1

CRL-A23

10% Coating by Weight of
Ethylcellulose on Sodium Chloride

(b) CAP-Coated Sodium Chloride and Disodium Phosphate

A process previously observed as being capable of producing small particle encapsulations involves depositing the protective polymer, CAP, onto solid particles. Encapsulation is accomplished by the phase-separation of CAP from solution by adding a second polymer, which is incompatible with CAP, to the CAP solution. Sodium chloride and disodium phosphate encapsulations were prepared in this preliminary study and excellent laboratory processing yields were achieved using the CAP system. Again, particle size control was achieved by milling the crystalline salts in the encapsulation solvent prior to inducing encapsulation. Figure 2 illustrates the weight distribution curve for a sodium chloride encapsulation (CRL-A20). The mean-volume diameter, d_v, is 37.4 microns with a standard deviation, σ, of 7.5 microns. Sample CRL-A21, a replication of Sample CRL-A20, illustrates the reproducibility of the milling-encapsulation process. The mean volume diameter, d_v, of Sample CRL-A21 is 39.3 microns with σ being 9.7 microns. The weight distribution curve is shown in Figure 3. Illustrated in Figure 4 is the weight distribution curve of a typical batch of CAP-coated disodium phosphate (Sample CRL-A22). The mean volume diameter, d_v, is 21.0 microns with a standard deviation, σ, of 12.0 microns. These results, as in the case of ethylcellulose, demonstrate the feasibility of small particle encapsulations using the CAP system.

As stated previously, CAP is a water permeable polymeric coating material, and is water insoluble in neutral or acidic solutions. The polymeric acid does become soluble, however, in alkaline solutions via formation of the water-soluble salt form. Hence, the method of activation or release for CAP-coated sodium chloride was expected to be that of diffusion, and, therefore, similar in sorption behavior to ethylcellulose-coated hygroscopic materials. The

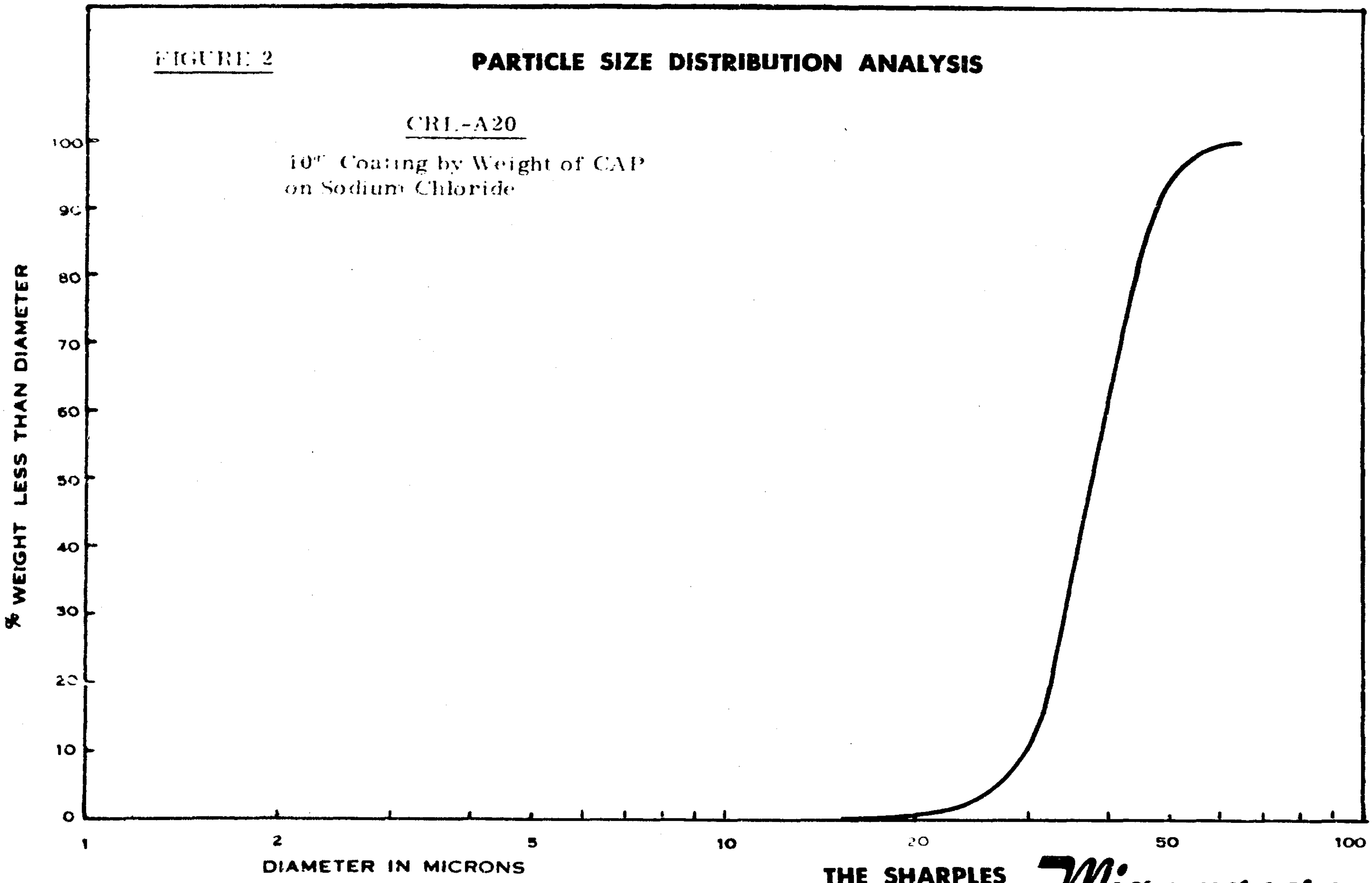

FIGURE 2

PARTICLE SIZE DISTRIBUTION ANALYSIS

CRL-A20

10% Coating by Weight of CAP
on Sodium Chloride

THE SHARPLES Micromerograph

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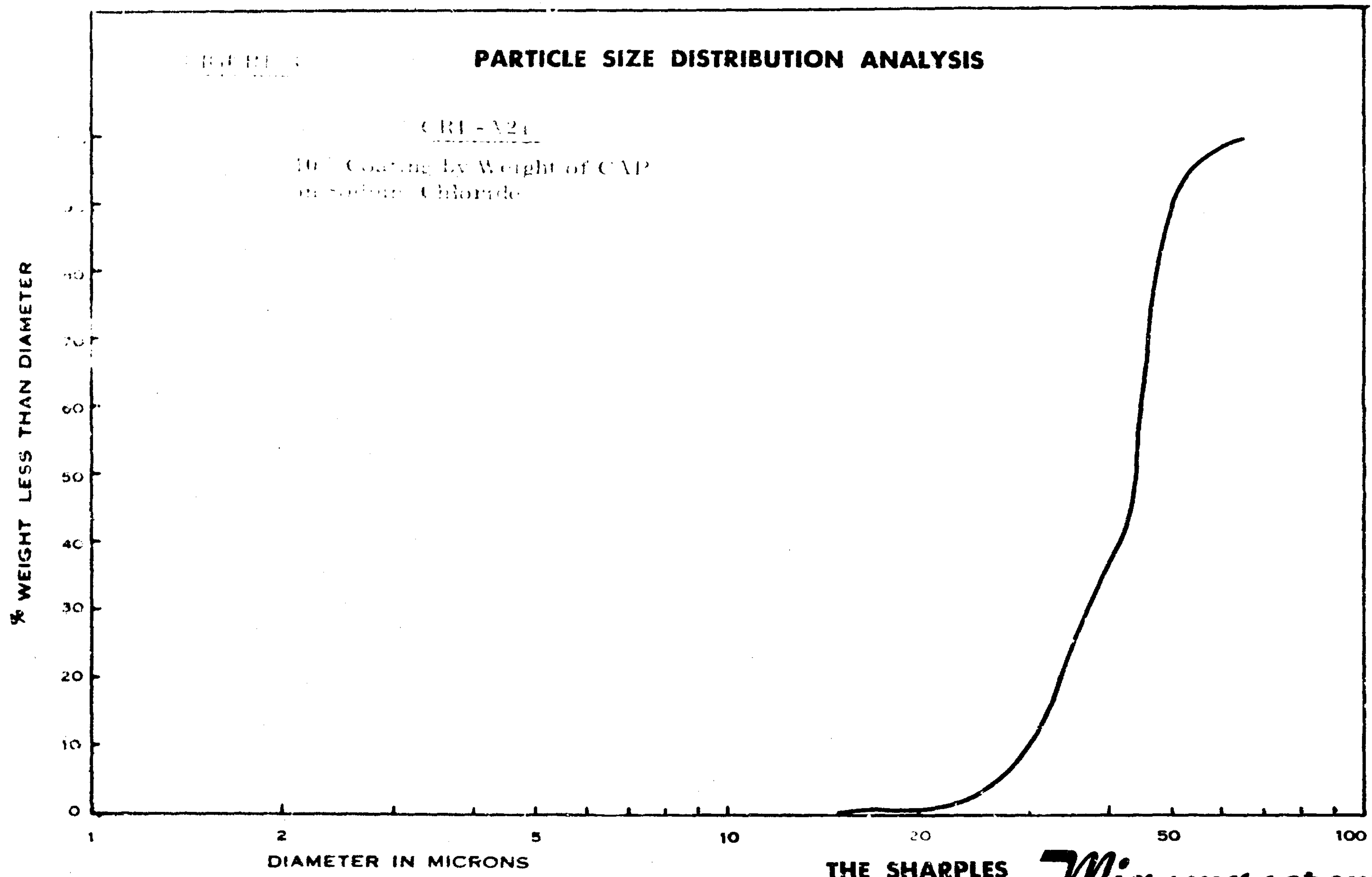
FIGURE 3



THE SHARPLES Micromerograph

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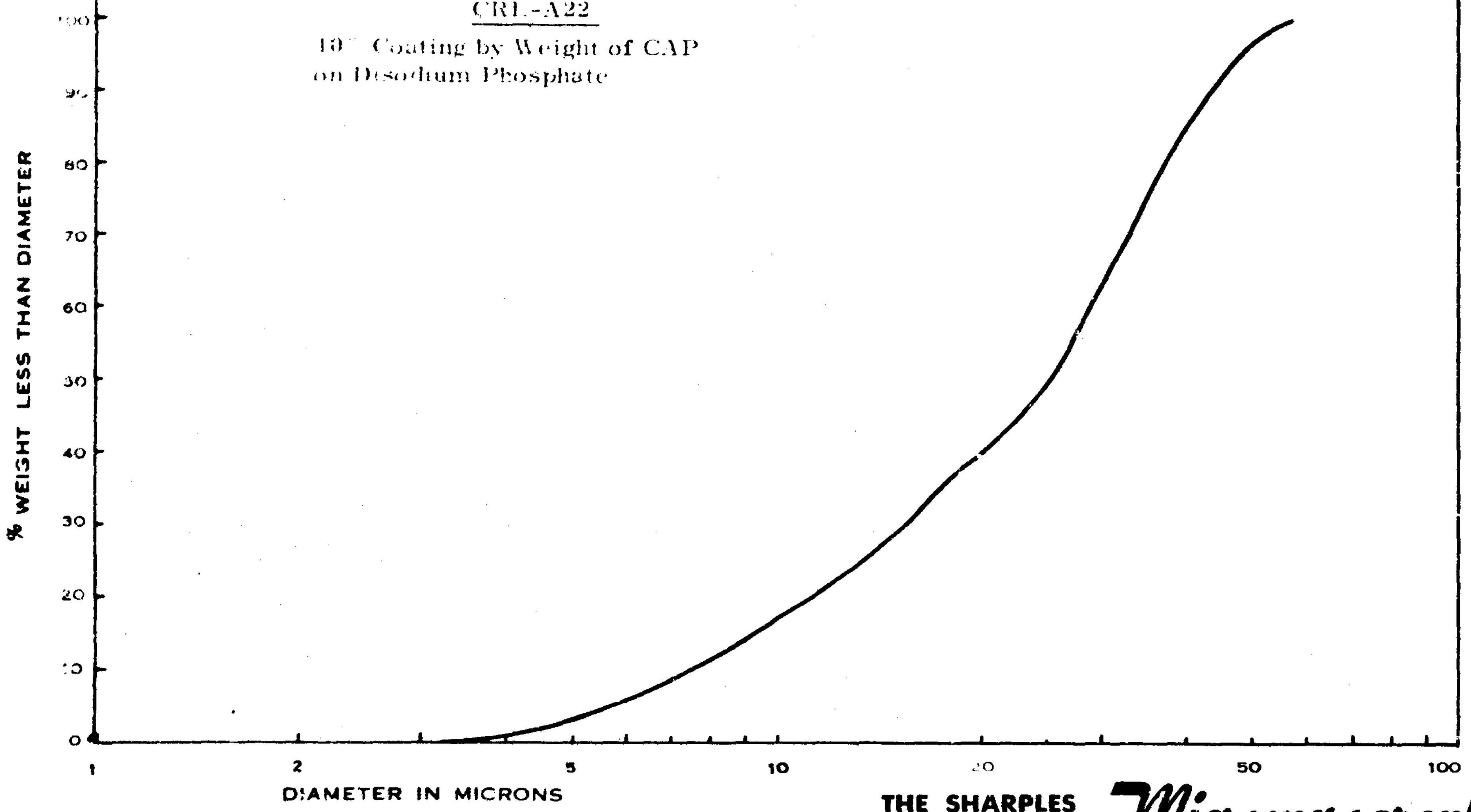
FIGURE 4

PARTICLE SIZE DISTRIBUTION ANALYSIS

CRL-A22

10% Coating by Weight of CAP
on Disodium Phosphate



THE SHARPLES **Micromerograph**
PARTICLE SIZE DISTRIBUTION ANALYSIS

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purpose of encapsulating the alkaline salt, dibasic sodium phosphate, was to provide an alkaline water droplet upon sorption of sufficient amounts of water, and thereby allow dissolution of the CAP coating. Also, thin films of CAP placed in mildly alkaline solutions swell immediately, and in so doing become infinitely more permeable to water and solute. It is this property which, hopefully, would provide quasi-instantaneous release or sorption properties.

3. Encapsulation of Sodium Chloride Solutions

The ability to encapsulate aqueous solutions of sodium chloride or other hygroscopic particles would eliminate the processing problem of milling crystalline materials to achieve the desired encapsulated particle size. The approach taken to achieve this capability was to encapsulate aqueous solutions of sodium chloride with ethylcellulose. Ethylcellulose is water permeable and will not retain water upon isolation of the capsules from the encapsulation media. Also, however, the polymer is not permselective to low molecular weight solute such as sodium chloride. Hence, the objective was to dehydrate capsules prior to isolation and to thereby induce *in situ* precipitation of the dissolved salt. Accomplishment of this would allow, perhaps, subsequent drying (solvent removal) of the capsules without undue loss of salt. Attempts to accomplish this were not successful with sodium chloride solutions. Limited *in situ* precipitation of sodium chloride within the encapsulated droplets was achieved, and excessive loss of salt during the dehydration processes occurred. The substitution of a highly water soluble salt for the moderately soluble sodium chloride could yield distinct advantages to this encapsulation concept and perhaps would show promise. However, the concept of encapsulated aqueous solutions was abandoned as a feasible approach for accomplishing the objectives of this particular program.

B. DEVELOPMENT OF ETHYLCELLULOSE-ENCAPSULATED SODIUM CHLORIDE

1. Encapsulation Process

It was decided to place emphasis on the process development of ethylcellulose encapsulated sodium chloride, until, at least, it could be shown that the diffusion-based particle growth was inappropriate for cloud seeding particles. The selection of ethylcellulose, rather than CAP for sodium chloride, was made because of processing considerations relative to the eventual objective of producing field test quantities of encapsulated materials. Also, it was found that a commercial grade of finely divided sodium chloride was amenable to small particle encapsulation processing. This allowed concentrating the effort on encapsulation, per se, and no further consideration was given to the preparatory grinding or milling aspects.

Using the Diamond Crystal Salt Company's sodium chloride, Microsized G-95 grade, encapsulation studies revealed that an effective and efficient encapsulation system was possible using cyclohexane as the polymer solvent or manufacturing vehicle. Using a particular grade of ethylcellulose (Ethocel, Standard 100 Grade, Dow Chemical Company), small encapsulated agglomerates could readily be achieved. The agglomerates could be prepared in the desired 20-80 micron range, and, in addition, the encapsulating-agglomerating process gave a product which was void, essentially, of fines. The particle size distributions of sodium chloride particles, before and after ethylcellulose encapsulation, is given in Figure 5. Also, typifying the process are photomicrographs given in Figure 6.

Attempts to optimize the process relative to processing yields per unit operation revealed an important characteristic. It was found that the particle size of the agglomerates varied inversely, within certain limits, with the ethylcellulose concentration employed. The

FIGURE 5 Particle Size (Weight) Distributions of Unencapsulated and Encapsulated Sodium Chloride

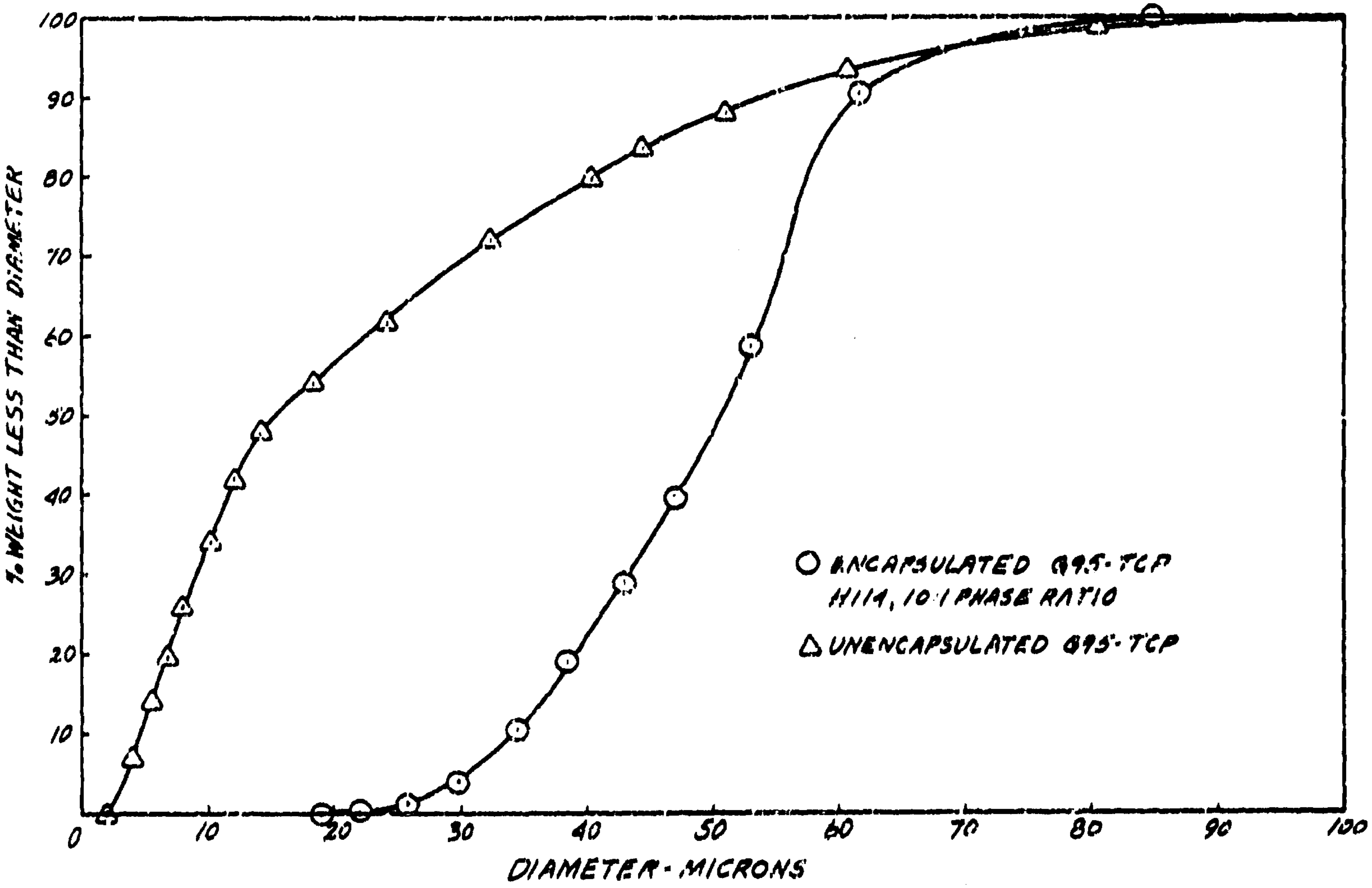
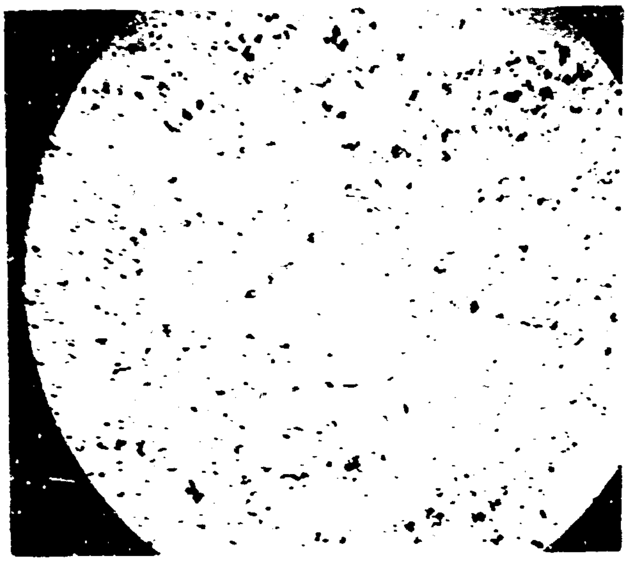
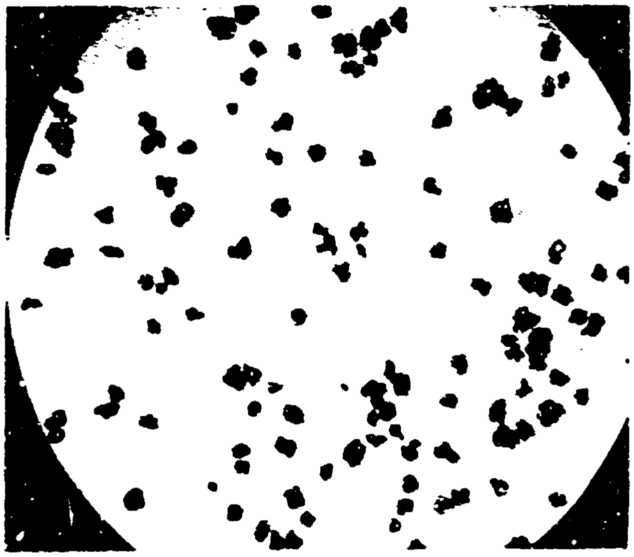


FIGURE 6

PHOTOMICROGRAPHS OF SODIUM CHLORIDE PARTICLES (LARGE SQUARE = 70 MICRONS)



Before Encapsulation



After Encapsulation

relationship is depicted in Figure 7. The importance of this aspect was two-fold; namely, it provided an added means of varying the size of the encapsulated agglomerates, and it allowed small particle encapsulation to be accomplished efficiently in terms of product yield per unit operation.

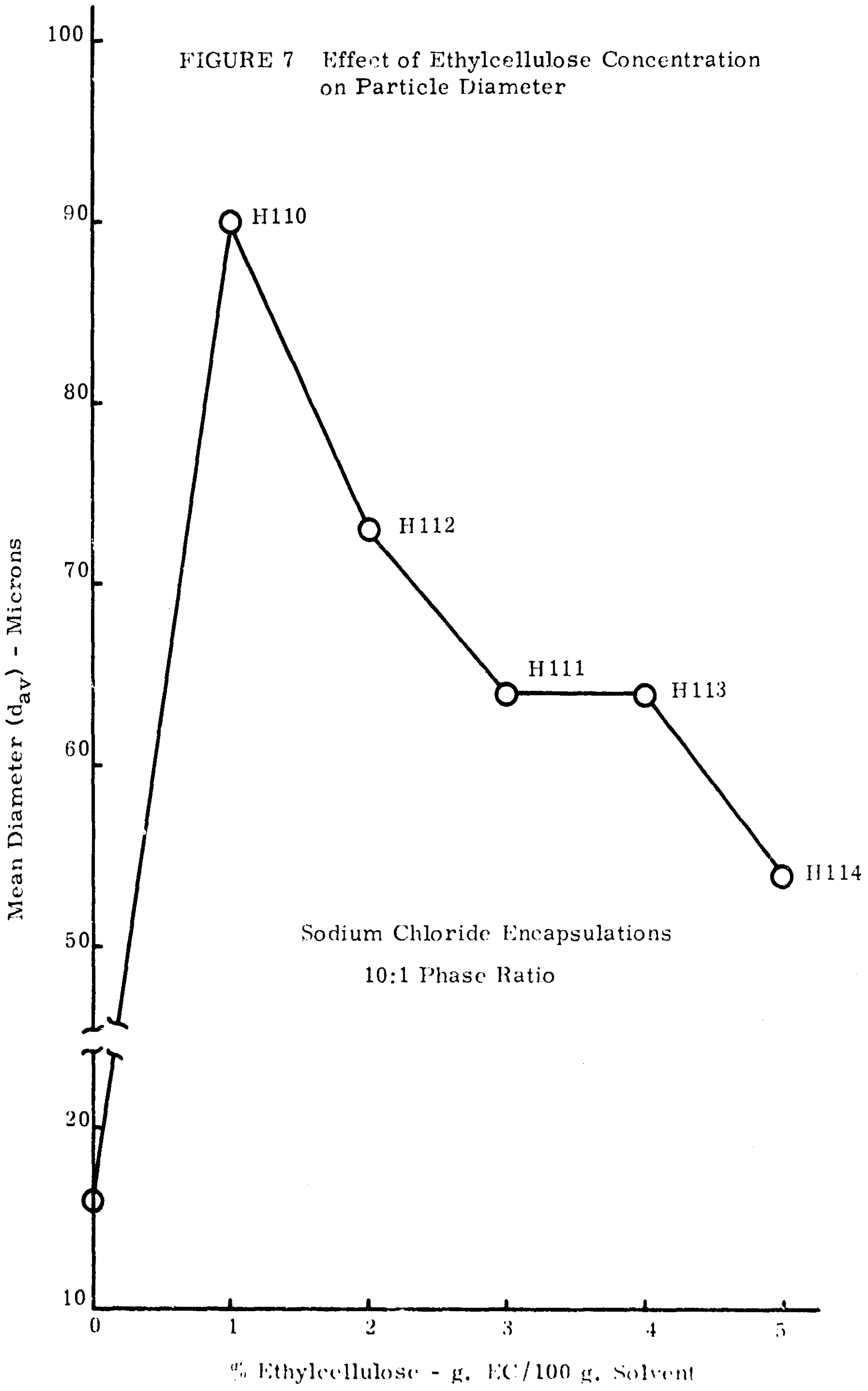
The relatively simple encapsulation process, with respect to materials and processing operations, is indicated in the schematic diagram presented in Figure 8.

2. Water Sorption Properties

(a) Gravimetric Method

A laboratory test method for comparing the water sorption characteristics of encapsulated and unencapsulated hygroscopic particles was developed. The instrumental method was designed around the capabilities of a DuPont Thermal Gravimetric Analyzer (TGA) and Differential Thermal Analyzer (DTA), Models 950 and 900, respectively. A sorption sample cell was fabricated and fitted to the electrobalance portion of the TGA unit. The cell was constructed to allow the continuous flow of a humid atmosphere over a sample mounted on the balance beam. Operating the DTA and TGA isothermally, allows the monitoring of sample weight versus time. The dynamic, humid environment is produced by bubbling compressed air through water in a thermostated chamber; a constant air velocity being maintained via a flow meter. The humidified atmosphere is then passed through the sorption cell containing the sample. The sorption cell is jacketed and is maintained at a temperature less than that of the incoming atmosphere. The humidity of the atmosphere is monitored with wet-dry Iron-Constantan thermocouples mounted in the atmosphere exit of the cell. The thermocouple emf (0°C reference junction) are recorded versus time using a two-pen strip recorder. The wet-dry temperatures are converted from the thermocouple millivolt readings. A schematic diagram of the apparatus is shown in Figure 9.

FIGURE 7 Effect of Ethylcellulose Concentration on Particle Diameter



Sodium Chloride Encapsulations

10:1 Phase Ratio

FIGURE 8

ENCAPSULATION PROCESSING STEPS

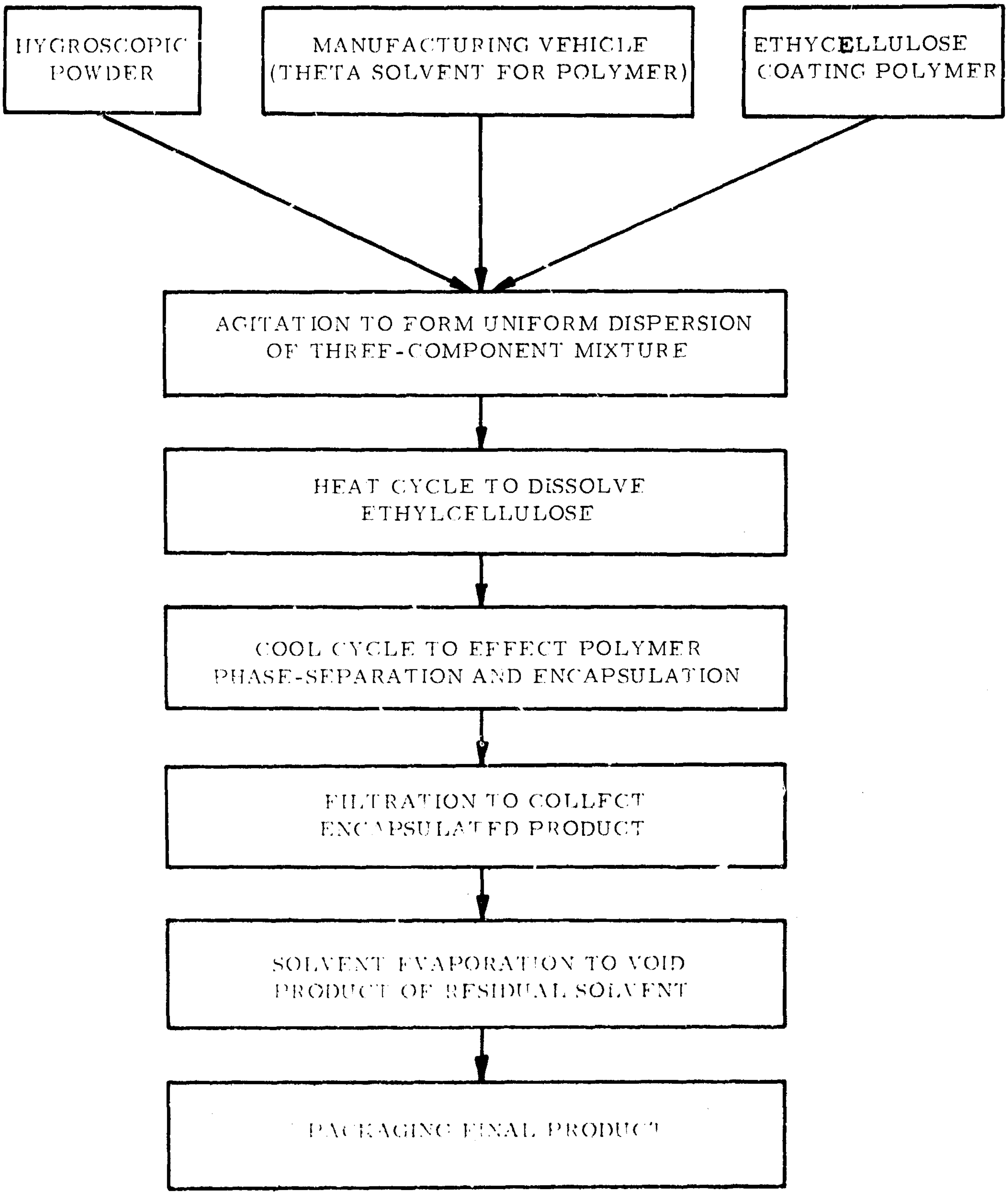
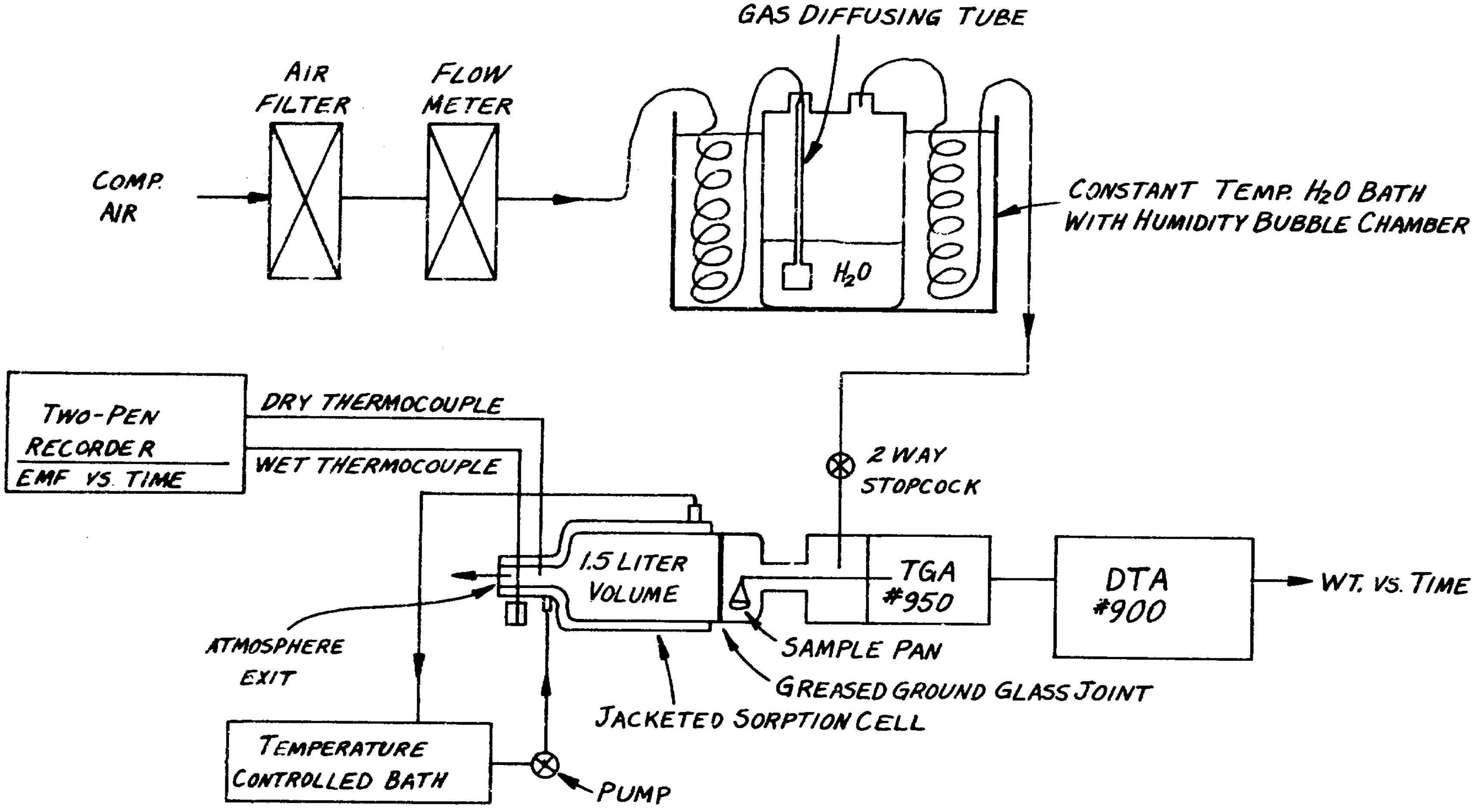


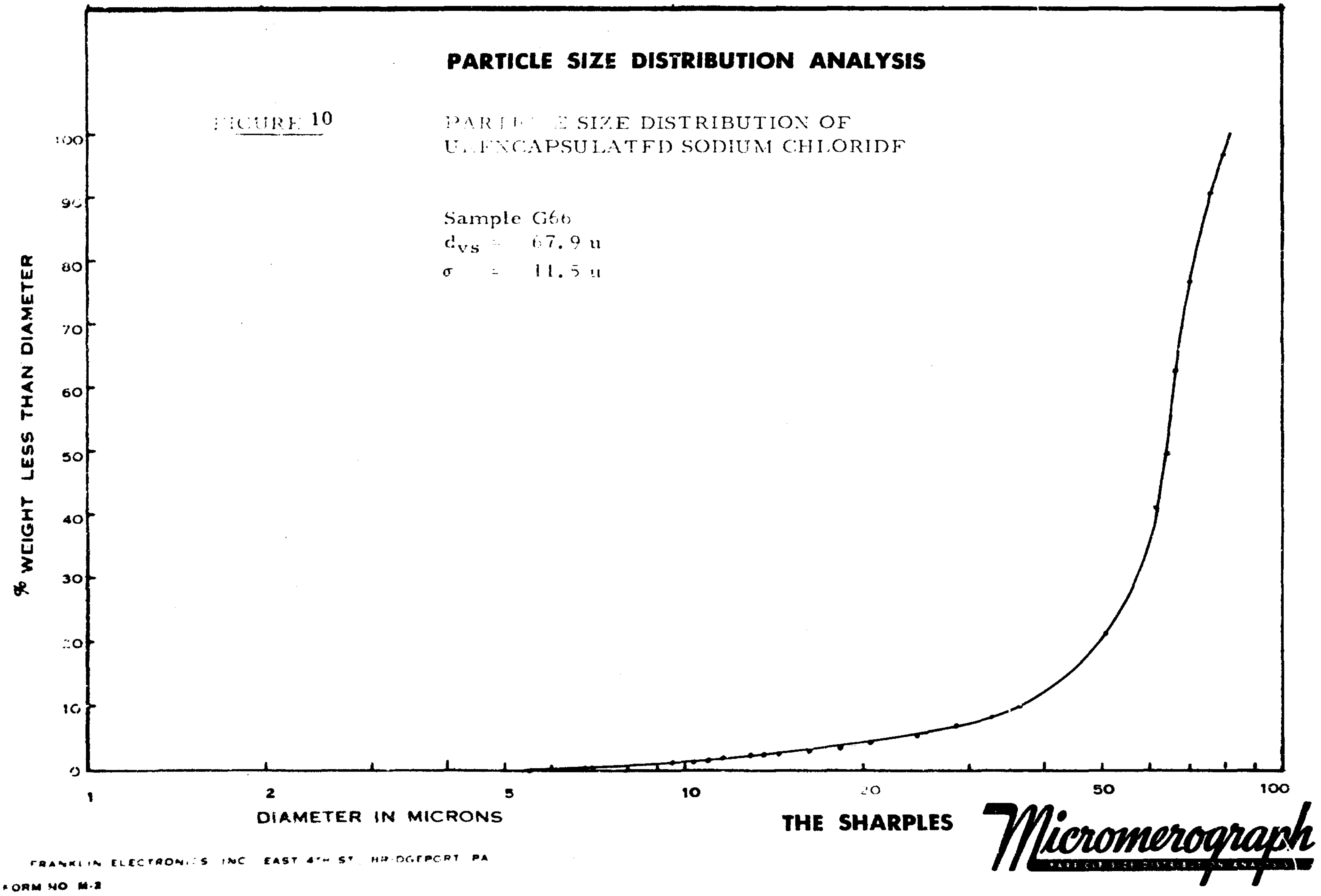
FIGURE 9 SCHEMATIC DIAGRAM OF ISOTHERMAL SORPTION APPARATUS



In practice, after extensive calibration, adjustment and technique development, reproducible results were achieved by employing the following method. An aluminum balance pan (20 mm. diameter) is placed on the balance beam and tared. The tared pan is removed and carefully loaded by dusting on the sample. Sample quantities of less than 1.5 milligrams are essential in achieving adequate dispersion of the particles. The loaded pan is replaced to the balance beam in the sorption cell, and the sample weight is recorded. The sample is then subjected to the dynamic, humid atmosphere and the weight is recorded versus time. The atmosphere velocity is maintained at 2 l./min. The wet-dry bulb temperatures are maintained at 20.0 ± 0.1°C giving a relative humidity of 96-100%. The recorded sample weight is corrected for a buoyance effect produced by the atmosphere flow and the weight gain (%) is calculated with respect to time (minutes).

A comparison of the sorption properties determined gravimetrically of unencapsulated sodium chloride material and an encapsulated sample (CRL-A102) are given in Tables I and II. The unencapsulated material was sieved to yield comparable particle size, and the micromerographic particle size distributions of the respective materials are shown in Figures 10 and 11. The results of the sorption study are shown graphically in Figure 12. The sorption data were utilized to calculate the diameter of the growing (average) particle versus time, and the results are given in Tables III and IV. Graphical treatments of the data are shown in Figures 13-15.

The gravimetric data show the water sorption properties of encapsulated sodium chloride to be comparable to the unencapsulated material. However, the calculated particle growth



PARTICLE SIZE DISTRIBUTION ANALYSIS

FIGURE 10

PARTICLE SIZE DISTRIBUTION OF
UNENCAPSULATED SODIUM CHLORIDE

Sample G66
d_{vs} = 67.9 u
σ = 11.5 u

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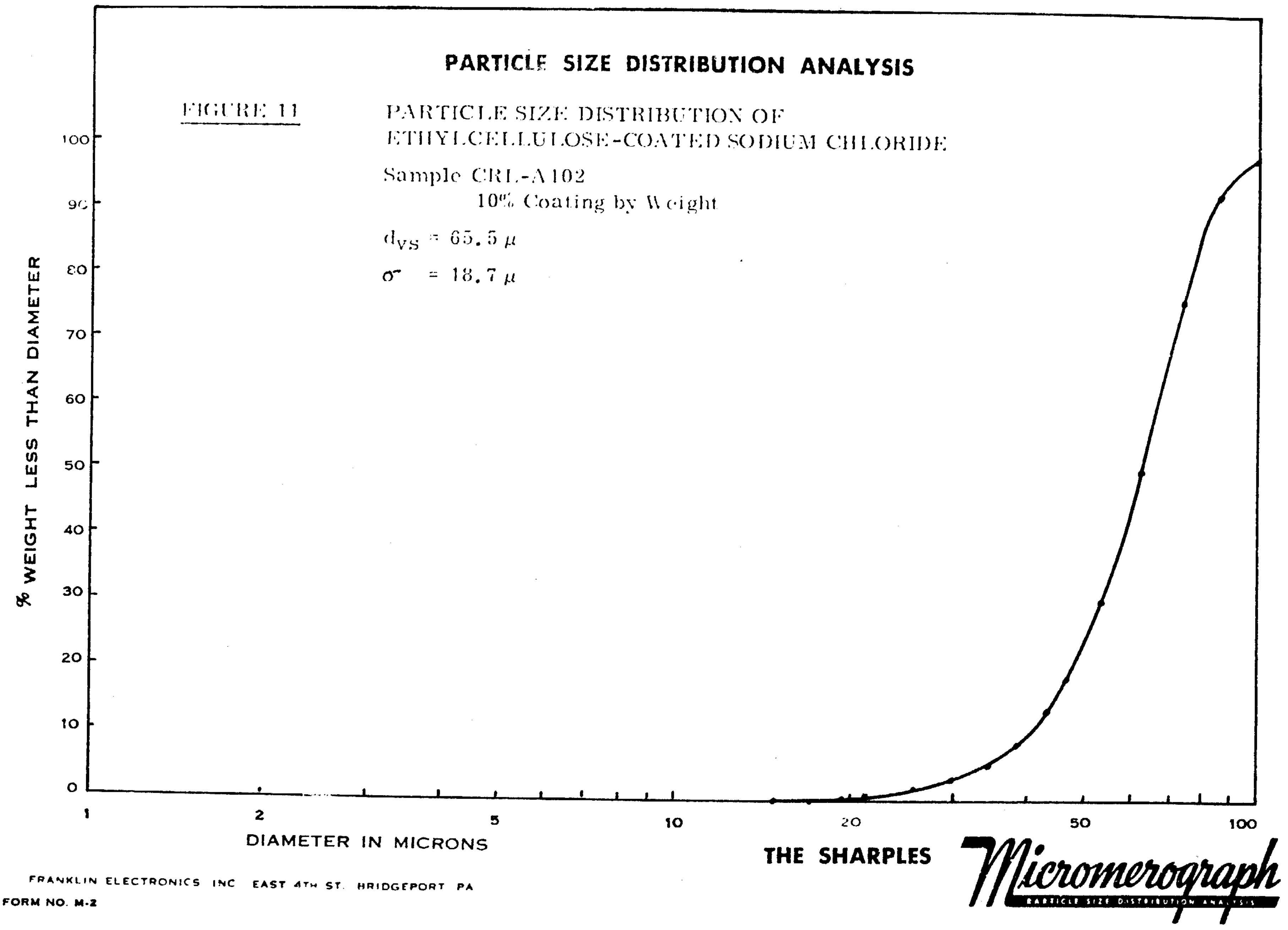


PARTICLE SIZE DISTRIBUTION ANALYSIS

FIGURE 11 PARTICLE SIZE DISTRIBUTION OF ETHYLCELLULOSE-COATED SODIUM CHLORIDE

Sample CRL-A102
10% Coating by Weight

$d_{vs} = 65.5\ \mu$

$\sigma = 18.7\ \mu$

TABLE I

Water Sorption of Unencapsulated NaCl at 100% RH, 20°C

Sample: G-66 Unencapsulated NaCl Control
Mean Volume-Surface Diameter, $d_{vs} = 67.9\ \mu$
Standard Deviation, $\sigma = 11.9\ \mu$

Determination	1	2	3	4	5	Avg.	d
Initial Sample Wt. (mg)	0.84	0.80	0.68	1.03	1.02	0.87	
Time/% Wt. Gain							
2.5 Min.	48%	30%	74%	42%	51%	49%	11
5.0	88	88	126	81	88	94	13
10.0	162	155	219	143	147	165	21
15.0	224	217	303	201	216	232	20
20.0	290	265	366	261	281	293	20
25.0	343	312	418	306	343	344	21
30.0	388	355	453	337	397	386	32
35.5	433	378	485	368	448	422	39
40.0	476	405	512	397	495	457	45
45.5	514	430	534	422	540	488	39
50.0	557	448	556	453	580	519	54
55.0	590	468	574	483	607	544	55
60.0	617	482	591	508	620	564	54

TABLE II

Water Sorption of Encapsulated NaCl at 100% RH, 20°C

Sample: CRL-A102 Ethylcellulose-Coated NaCl
Mean Volume-Surface Diameter, $d_{vs} = 65.5\ \mu$
Standard Deviation, $\sigma = 18.7\ \mu$

Determination	1	2	3	4	5	Avg.	d
Initial Sample Wt. (mg)	0.77	0.90	1.10	0.98	0.91	0.93	--
Time/% Wt. Gain							
2.5 Min.	67%	57%	57%	59%	60%	60%	3
5.0	122	121	108	112	107	114	6
10.0	245	248	214	214	198	224	18
15.0	332	358	285	297	278	314	29
20.0	399	440	386	371	344	388	25
25.0	482	513	454	428	397	455	31
30.0	565	569	510	478	442	515	42
35.0	606	618	555	514	476	554	41
40.0	656	658	606	542	506	594	56
45.0	697	704	661	571	528	632	65
50.0	736	742	701	606	550	667	72
55.0	757	787	723	633	560	692	73
60.0	788	804	755	659	572	716	80

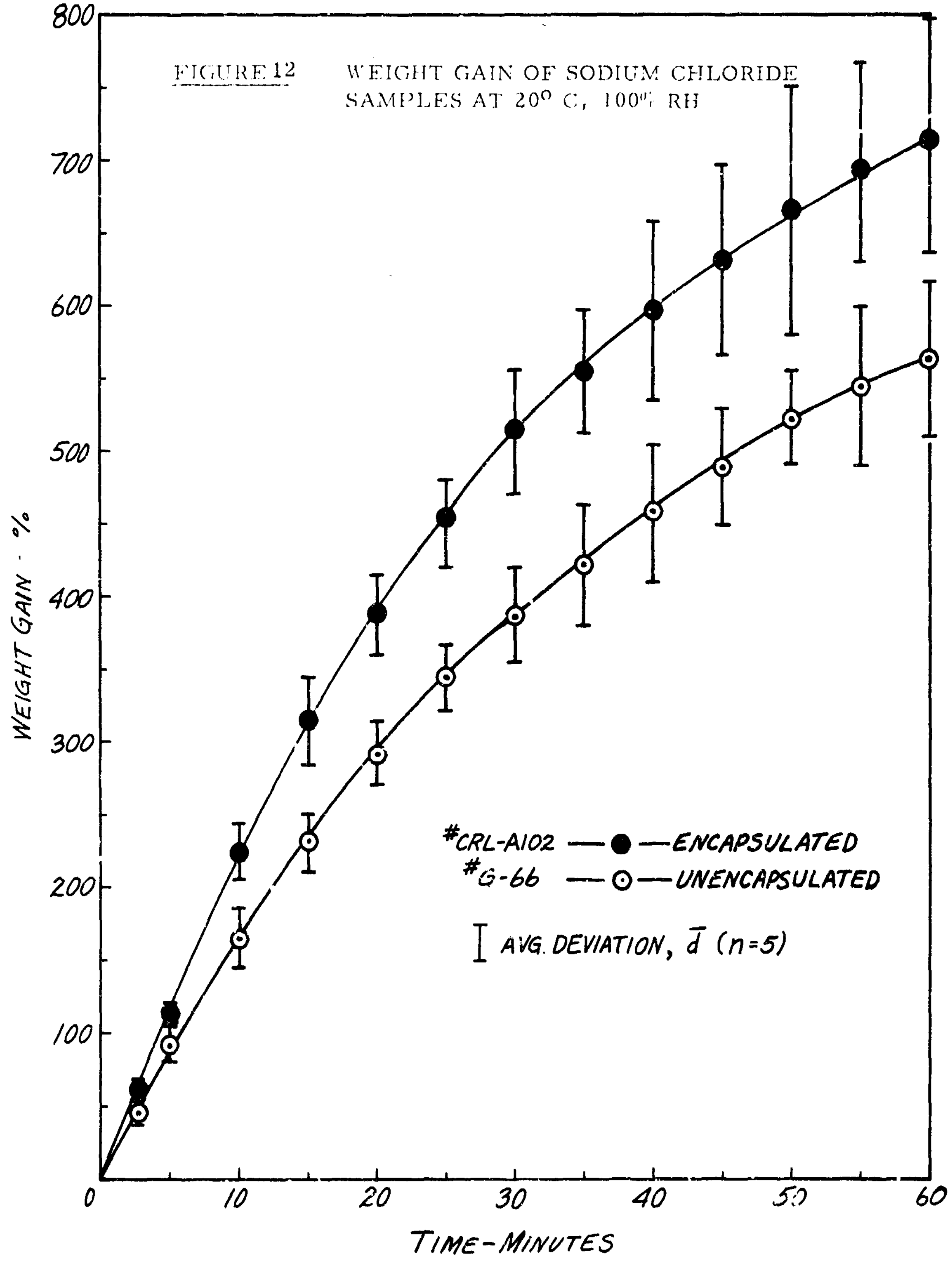


FIGURE 12 WEIGHT GAIN OF SODIUM CHLORIDE SAMPLES AT 20° C, 100% RH

TABLE III

Calculation of Diameters for Growing Particles of Unencapsulated Sodium Chloride

Sample No. G-66

Known:
Avg. Sample Wt. = 8.7×10^{-4} g.
$d_{vs} = 67.9\ \mu$
$\rho = 2.16$ g/cm^3 at 20°C*
$C_3 = 0.358$ g NaCl/g. H_2O at 20°C = 26.4 wt. %**
Particle Wt. = 3.54×10^{-7} g.
No. Particles = 2.46×10^3
Temp. = 20.0° C

Assumed: Instantaneous dissolution, instantaneous solute diffusion and spherical particles.

Time (min.)	Avg. Wt. Gain (%)	Wt. of H_2O (g/sample)	Wt. of H_2O (g/particle)	Wt. of Solute (g/particle)	Solute Conc. (wt. %)	Solution*** Density (g/cm^3)
2.5	49	5.58×10^{-4}	1.73×10^{-7}	0.62×10^{-7}	26.4	1.202
5.0	94	10.6	3.32	1.19	26.4	1.202
10.0	165	20.8	5.86	2.10	26.4	1.202
15.0	232	29.2	8.24	3.10	26.4	1.202
20.0	293	36.2	10.4	3.54	25.5	1.194
25.0	344	42.3	12.2	3.54	22.5	1.171
30.0	386	47.8	13.7	3.54	20.5	1.155
35.0	422	51.4	14.9	3.54	19.2	1.142
40.0	457	55.2	16.2	3.54	18.0	1.134
45.0	488	58.6	17.3	3.54	17.0	1.126
50.0	519	61.9	18.4	3.54	16.2	1.119
55.0	544	64.3	19.3	3.54	15.5	1.114
60.0	564	66.5	20.0	3.54	15.0	1.111

TABLE III (Cont.)

Time (min.)	Solution Wt. (g/particle)	Solution Vol. (cm^3/particle)	NaCl (solid) Remaining (g/particle)	Vol. NaCl (solid) (cm^3/particle)	Total Vol. (cm^3/particle)	Particle Diameter (μ)
2.5	2.35×10^{-7}	1.95×10^{-7}	2.92×10^{-7}	1.35×10^{-7}	3.30×10^{-7}	85.7
5.0	4.52	3.75	2.35	1.09	4.84	97.4
10.0	7.96	6.62	1.44	0.68	7.30	112
15.0	11.2	9.32	0.59	0.27	9.57	122
20.0	14.1	11.7	0.00	0.00	11.7	131
25.0	16.4	14.0	0.00	0.00	14.0	139
30.0	18.6	16.1	0.00	0.00	16.1	145
35.0	20.2	17.7	0.00	0.00	17.7	150
40.0	22.0	19.2	0.00	0.00	19.2	154
45.0	23.5	20.9	0.00	0.00	20.9	159
50.0	25.0	22.3	0.00	0.00	22.3	162
55.0	26.2	23.5	0.00	0.00	23.5	165
60.0	27.2	24.4	0.00	0.00	24.4	167

*Determined experimentally

**From "International Critical Tables"

***From "Handbook of Chemistry and Physics", 48th ed.

TABLE IV

Calculation of Diameters for Growing Particles of Encapsulated Sodium Chloride

Sample No. CRL-A102

Known:
Avg. Sample Wt. = 9.3×10^{-4} g.
$d_{vs} = 65.5\ \mu$
$\rho = 2.00\ g/cm^3$*
Particle Wt. = 2.94×10^{-7} g.
NaCl wt. = (2.94×10^{-7}) (90%) = 2.65×10^{-7} g.
C_s = 0.358 g NaCl/g. H_2O at 20°C = 26.4 wt. %**
No. of particles = 3.16×10^3
Temp. = 20.0°C

Assumed: Instantaneous dissolution, instantaneous solute diffusion, and spherical particles

Time (min.)	Avg. Wt. Gain (%)	Wt. of H_2O (g/sample)	Wt. of H_2O (g/particle)	Wt. of Solute (g/particle)	Solute Conc. (wt. %)	Solution *** Density (g/cm^3)
2.5	60.2	5.58×10^{-4}	1.77×10^{-7}	0.634×10^{-7}	26.4	1.202
5.0	114	10.6	3.36	1.20	26.4	1.202
10.0	224	20.8	6.57	2.34	26.4	1.202
15.0	314	29.2	9.25	2.65	22.6	1.172
20.0	388	36.2	11.5	2.65	18.8	1.141
25.0	455	42.3	13.4	2.65	16.5	1.123
30.0	515	47.8	15.2	2.65	14.9	1.109
35.0	554	51.4	16.3	2.65	14.0	1.103
40.0	594	55.2	17.4	2.65	13.2	1.096
45.0	632	58.6	18.6	2.65	12.5	1.091
50.0	667	61.9	19.6	2.65	11.9	1.087
55.0	692	64.3	20.3	2.65	11.6	1.085
60.0	716	66.5	21.2	2.65	11.1	1.081

TABLE IV (Cont.)

Time (min.)	Solution Wt. (g/particle)	Solution Vol. (cm^3/particle)	NaCl (solid) Remaining (g/particle)	Vol. NaCl (solid) (cm^3/particle)	Total Vol. (cm^3/particle)	Particle Diameter (μ)
2.5	2.40×10^{-7}	1.99×10^{-7}	2.02×10^{-7}	0.927×10^{-7}	2.92×10^{-7}	82.2
5.0	4.56	3.79	1.45	0.67	4.46	94.8
10.0	8.91	7.42	0.31	0.14	7.56	112
15.0	11.9	10.2	0.00	0.00	10.2	125
20.0	14.1	12.4	0.00	0.00	12.4	133
25.0	16.1	14.3	0.00	0.00	14.3	140
30.0	17.8	16.1	0.00	0.00	16.1	145
35.0	18.9	17.3	0.00	0.00	17.3	149
40.0	20.1	18.4	0.00	0.00	18.3	150
45.0	21.2	19.4	0.00	0.00	19.4	155
50.0	22.2	20.4	0.00	0.00	20.4	157
55.0	23.0	21.2	0.00	0.00	21.2	159
60.0	23.9	22.1	0.00	0.00	22.1	162

*Determined experimentally

**From "International Critical Tables"

***From "Handbook of Chemistry and Physics", 48th Ed.

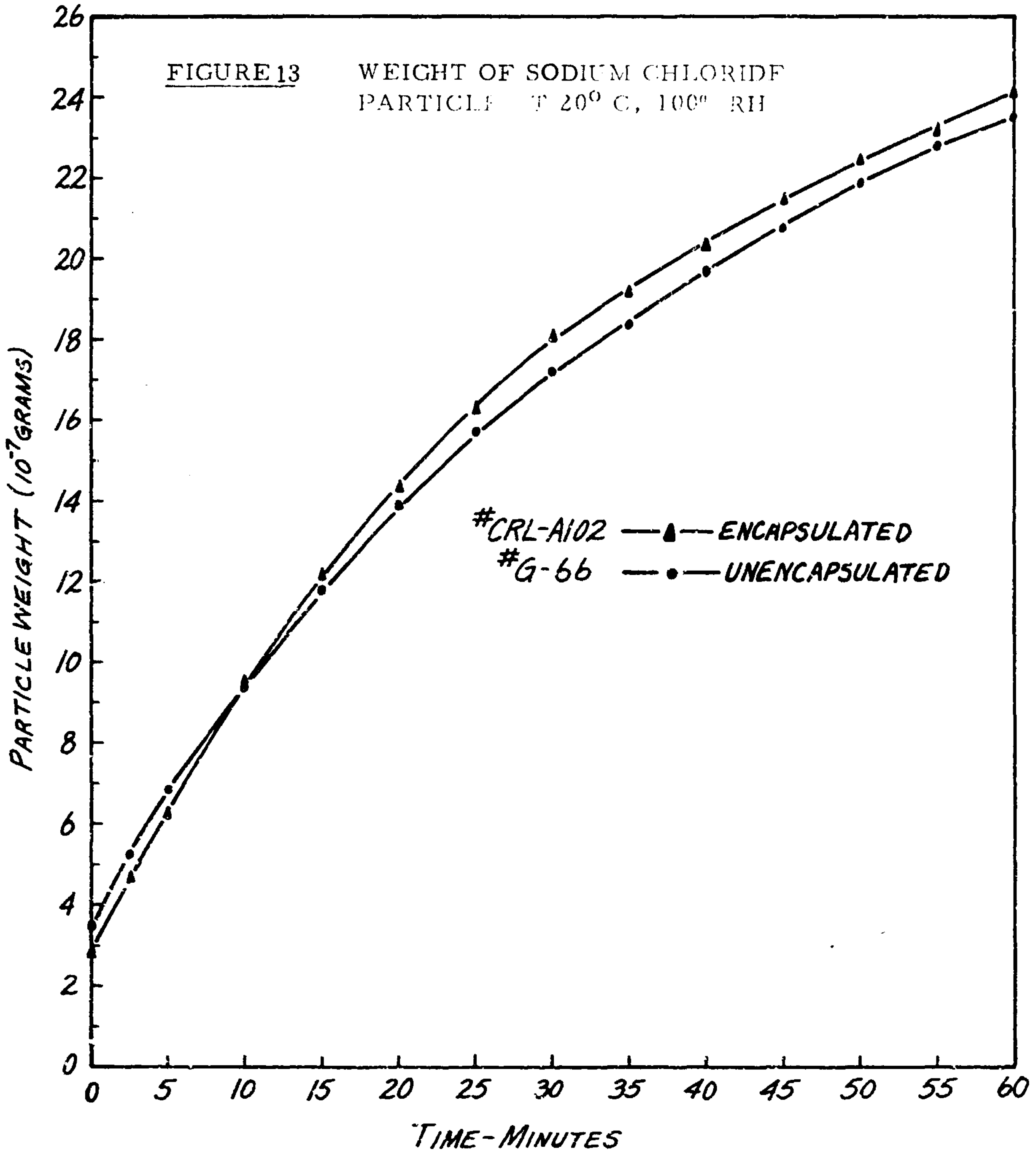


FIGURE 13 WEIGHT OF SODIUM CHLORIDE PARTICLE AT 20° C, 100% RH

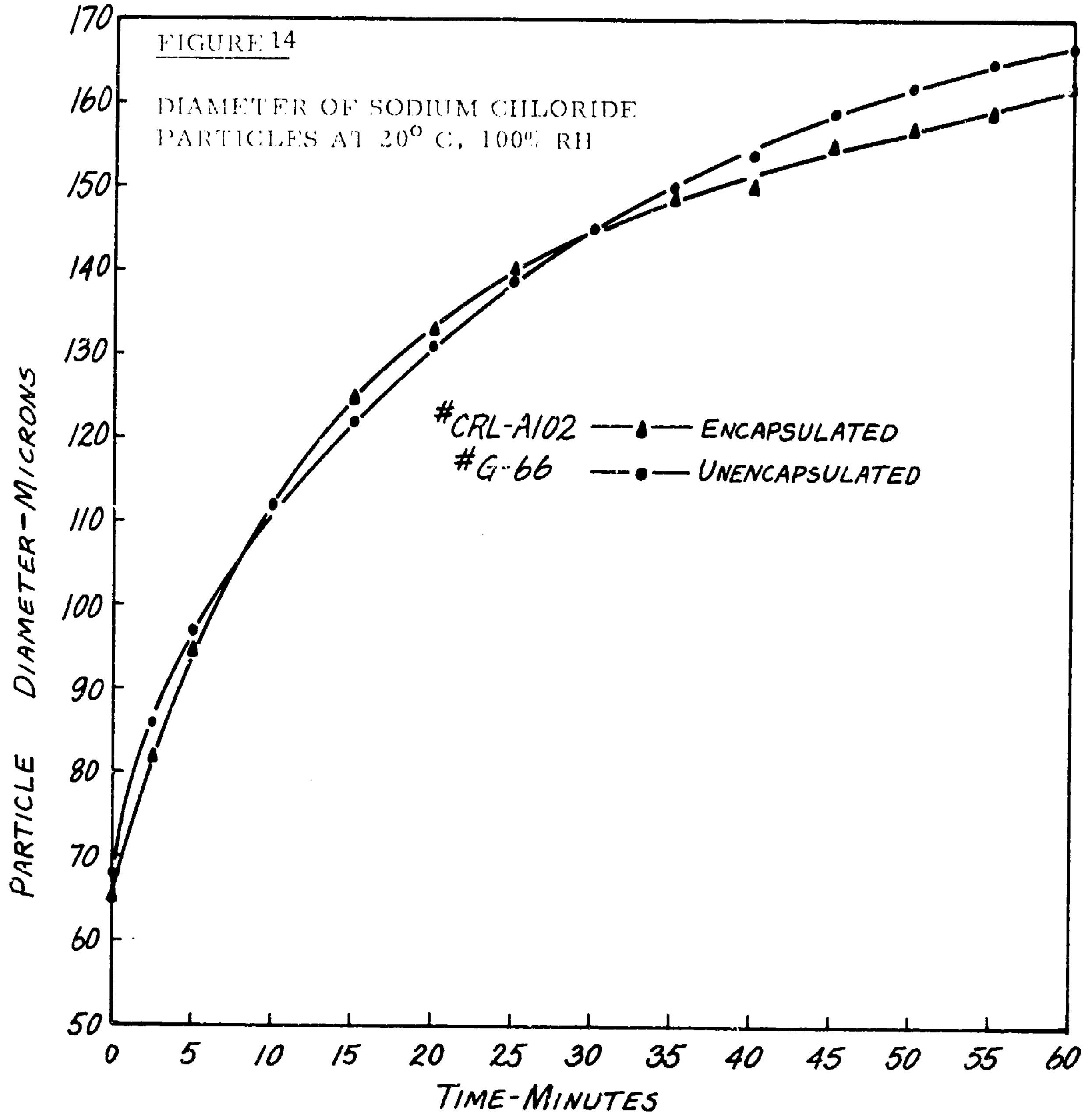
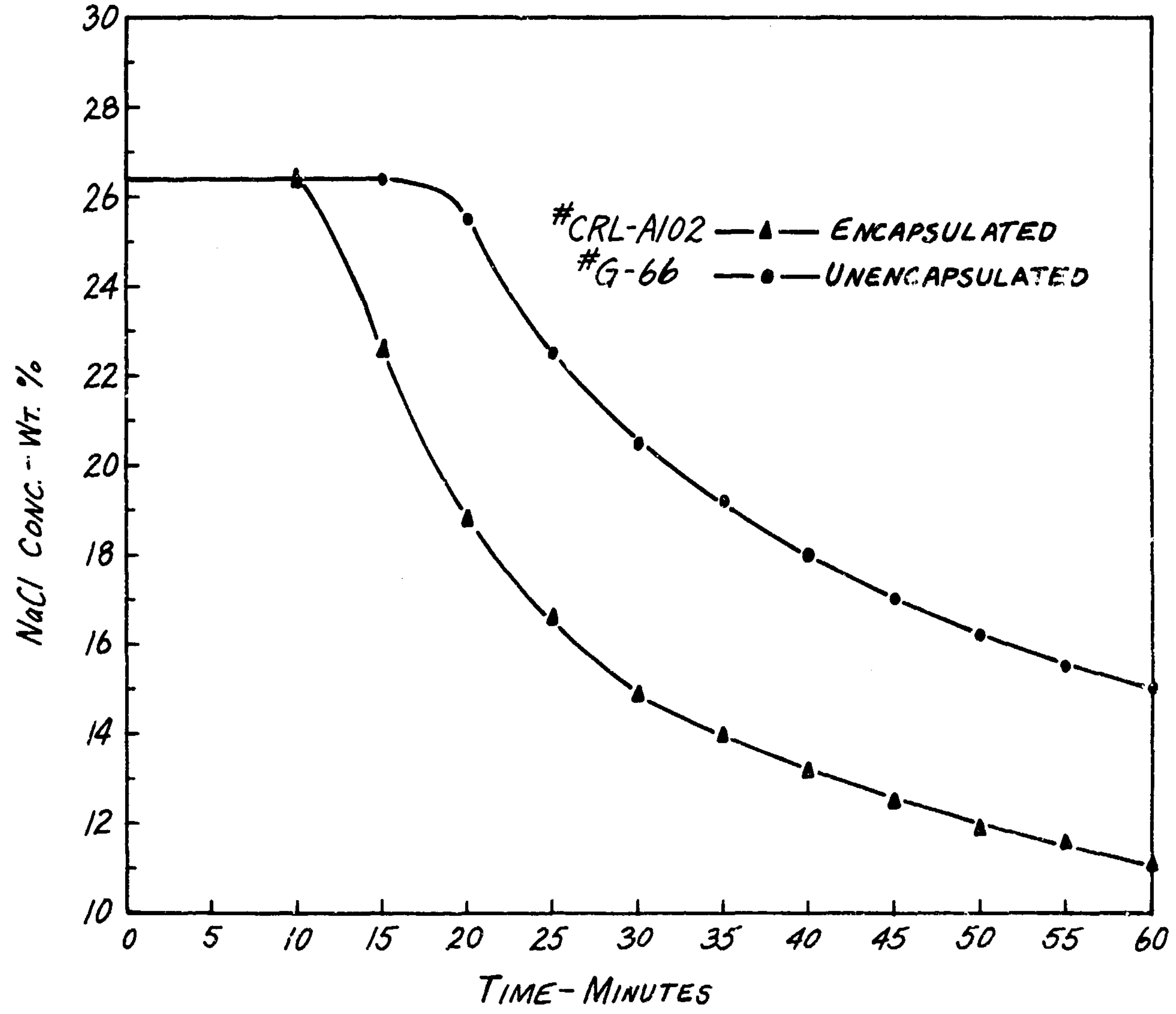


FIGURE 14

DIAMETER OF SODIUM CHLORIDE PARTICLES AT 20° C, 100% RH

FIGURE 15 SODIUM CHLORIDE SOLUTE CONCENTRATION OF PARTICLE AT 20°, 100% RH

Solubility, C_s = 26.4 wt. % @ 20° C.



characteristics were considerably retarded compared to the theoretical values supplied by AFCRL. The particle diameter versus time relationships were calculated from gravimetric data using a "pan" method whereby the average weight gain for several thousand particles was observed with each determination. It had been assumed that the gravimetric method would provide valid comparisons of the growth rates of unencapsulated and encapsulated salts, and that the data would be extrapolatable to atmospheric warm fog conditions. However, it was proposed to supplement the gravimetric data with optical measurements of particle growth incurred upon exposure of the particles to a humid (ca 100% RH) environment. The optical method employed and the results are described below.

(b) Optical Method

A version of the optical method employed by Ye Jia-Dong[1] was investigated. The simplified method consisted of the following: A single particle is mounted on a horizontally suspended, glass thread. The thread diameter is 10 microns. The sample is placed over 2 cc. of water contained in an 8 cc. humidity cell. The assembly is placed under a microscope and the particle diameter is determined optically. The chamber is closed with a glass cover-plate and the particle growth versus time is measured by photomicrography.

Using the optical method, 60-70 micron encapsulated (Sample CRL-A102) and unencapsulated sodium chloride particles displayed growth rates which corroborated the gravimetric data previously reported; namely, ethylcellulose-coated aggregates grow at rates comparable to those of unencapsulated crystals of the same dimensions. Also, the optical method produces rapid particle growth and better agrees with theoretical values. Figure 16 compares the particle growth curves obtained experimentally with the theoretical growth curve determined by AFCRL using a numerical model.

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[1] Ye Jia-Dong, "An Experimental Study on Artificial Condensation Nuclei," AFCRL-68-0629, Translations, No. 36, 1968.

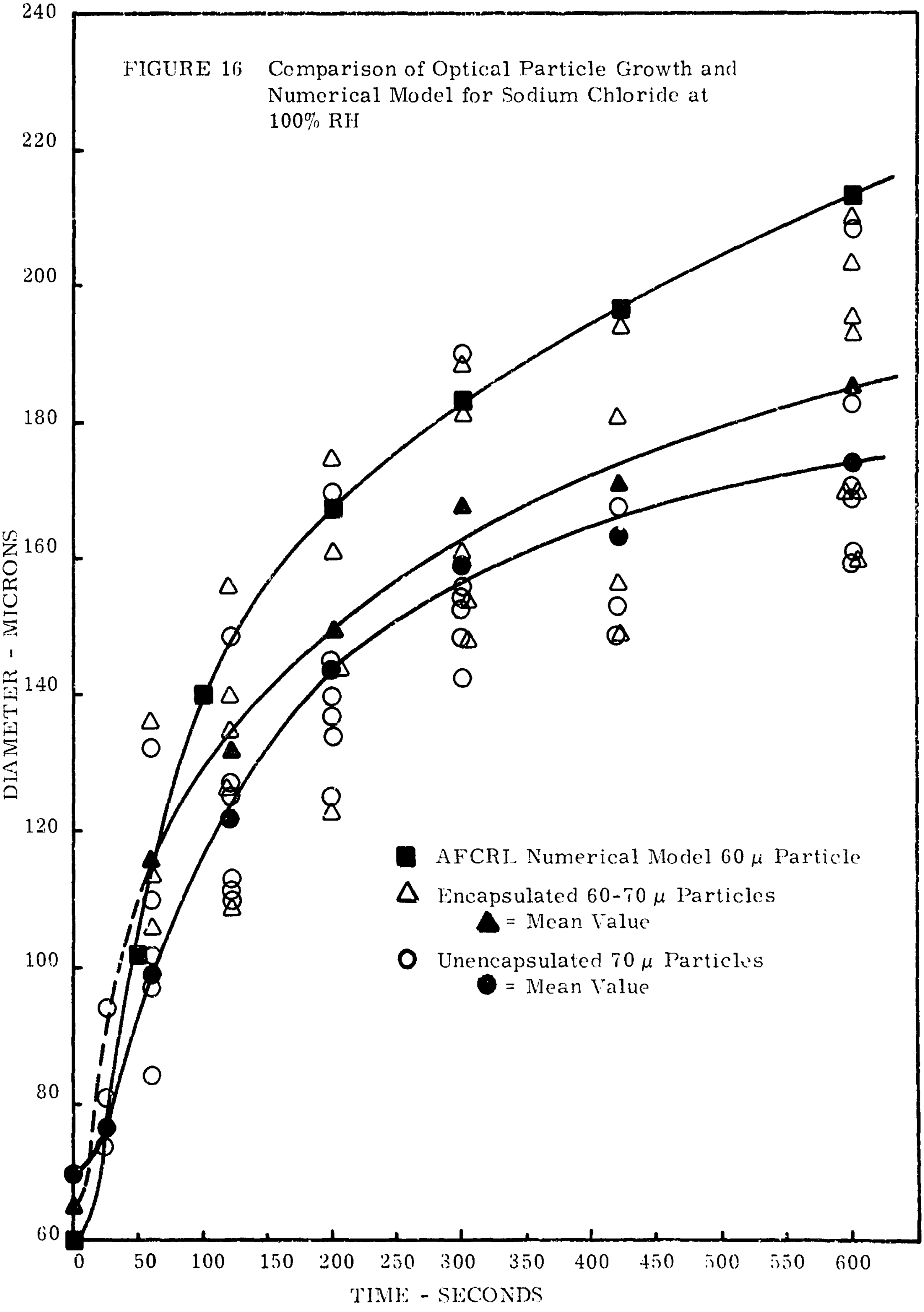


FIGURE 16 Comparison of Optical Particle Growth and Numerical Model for Sodium Chloride at 100% RH

Considering the results of the particle growth studies, the following discussion is offered.

The protective coating material, ethylcellulose, is a water-insoluble polymer, which when applied as a thin microencapsulation coating is permeable to water vapor, liquid water and dissolved solutes. Mass transport processes are accomplished by diffusion mechanisms operating across the polymeric membrane. Although the polymer is permeable, it would be expected that encapsulated hygroscopic particles would exhibit some degree of inhibited water sorption. The tests described above, however, showed the encapsulated agglomerates to be at least as sorptive as unencapsulated particles of comparable dimension. In some cases, water sorption appeared to be more rapidly accomplished with the encapsulated agglomerates. Explanation of these results involves the agglomerating aspect achieved by encapsulation and the intraparticle porosity of the resulting particles. The finely divided sodium chloride raw material has a specific surface of about 4,000 cm^2/cm^3. Encapsulation (and agglomeration) produces encapsulated particles having a specific surface of about 700 cm^2/cm^3. The effective surface of the sodium chloride, however, remains essentially that which it was prior to encapsulation, viz., 4,000 cm^2/cm^3. It is the hypothesis that the intraparticle surface of the encapsulated hygroscopic particles is offsetting to the water sorption inhibition incurred by the presence of the ethylcellulose coating.

3. Bulk Powder Properties

Attempts were made to utilize an angle of repose measurement to empirically evaluate the agglomerating tendencies of unencapsulated and encapsulated sodium chloride materials. The intent was to expose the bulk powders to a humid environment and compare the respective angles of repose before and after exposure. A variation of the funnel method for determining the angle was investigated. The method, completely satisfactory for the encapsulated materials (sodium chloride), was inoperative for the unencapsulated powders due to their excessive agglomerating tendencies when exposed to humid environments.

Another empirical test was developed, however, and satisfactory results were obtained. The method involved the following: Fifty cc. of bulk powder are measured into a 120 cc., wide-mouth bottle. The sample surface/volume ratio is 13 $cm^2/50\ cm^3$. The bottle is placed in a humidity chamber, maintained at 32°C, 90% RH, for 120 minutes. The bottle is then removed, capped and shaken vigorously by hand for 60 seconds. The sample is then placed on a 100 mesh (149 μ) sieve-pan combination. The sieve is gently shaken until the screen has obviously cleared of fine material. The two fractions are collected, dried and weighed. The weight percent retained on the screen is calculated and is utilized as a measure of the degree of agglomeration occurring.

Results typifying the experimental method are given below for two sodium chloride materials referenced previously.

Determination		G66-Unencapsulated Control		CRL-102A Ethyl-cellulose-Coated
1		27% Retained		1.8% Retained
2		26		1.5
3		25		1.4
4		26		2.0
Avg. % Retained	=	26%	=	1.7%

The above results demonstrate that the ethylcellulose encapsulation substantially decreased the agglomeration tendency of sodium chloride bulk powder when exposed to the conditions described above.

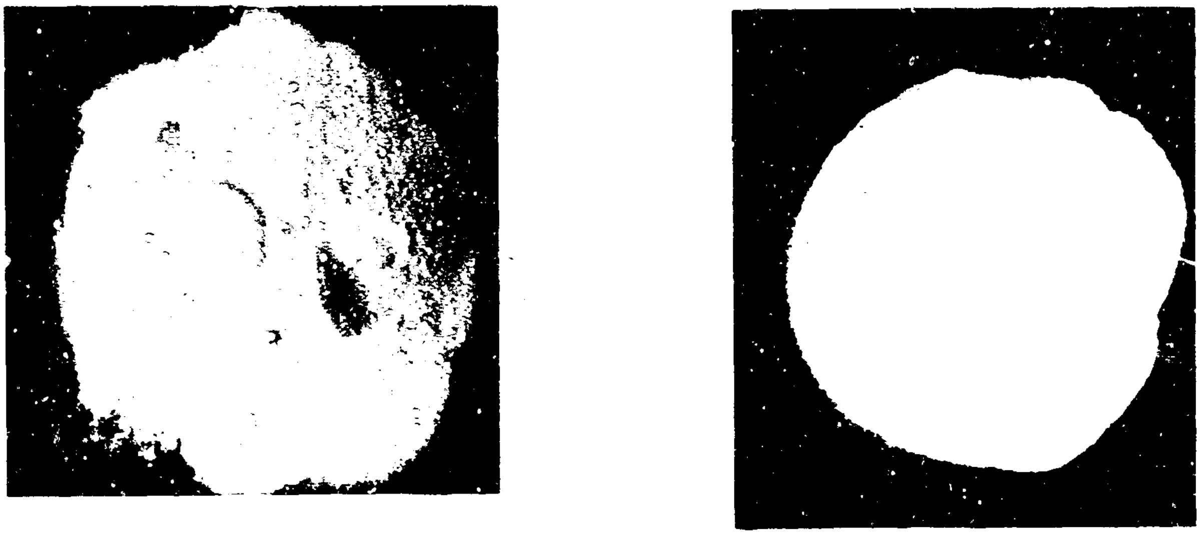
The photographs shown in Figure 17 illustrate the retained fractions of encapsulated and unencapsulated powders.

FIGURE 17

SODIUM CHLORIDE BULK POWDERS AFTER OPEN-CONTAINER EXPOSURE TO 32°C/90% RH FOR 120 MINUTES

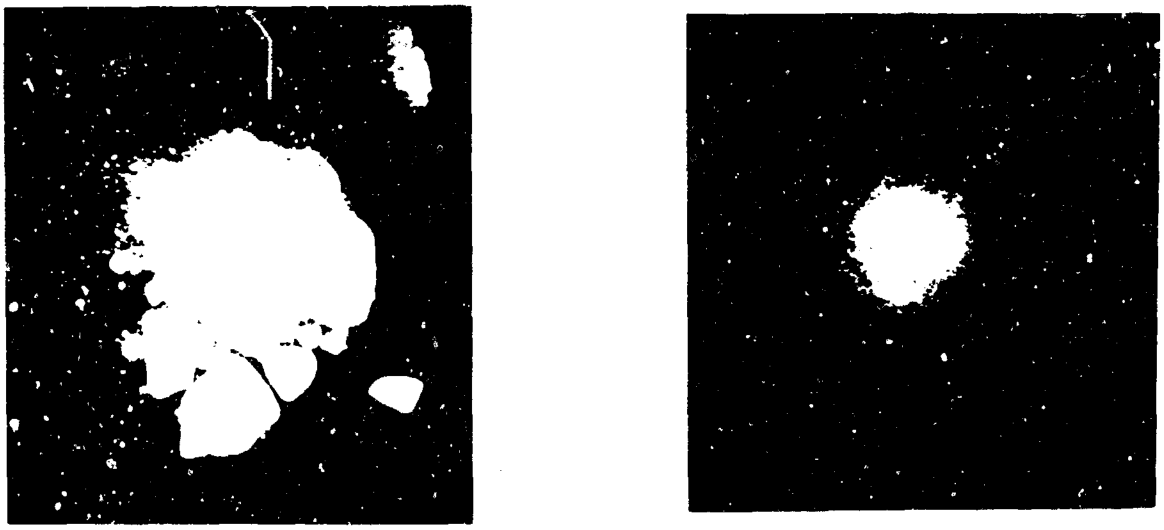
Unencapsulated Powder

Encapsulated Powder



Total Sample

Total Sample



Fraction Retained on
100 Mesh Screen
(26 wt. %)

Fraction Retained on
100 Mesh Screen
(2 wt. %)

C. TRANSLATION OF SODIUM CHLORIDE TECHNOLOGY TO UREA

1. Encapsulation Process

(a) Feasibility

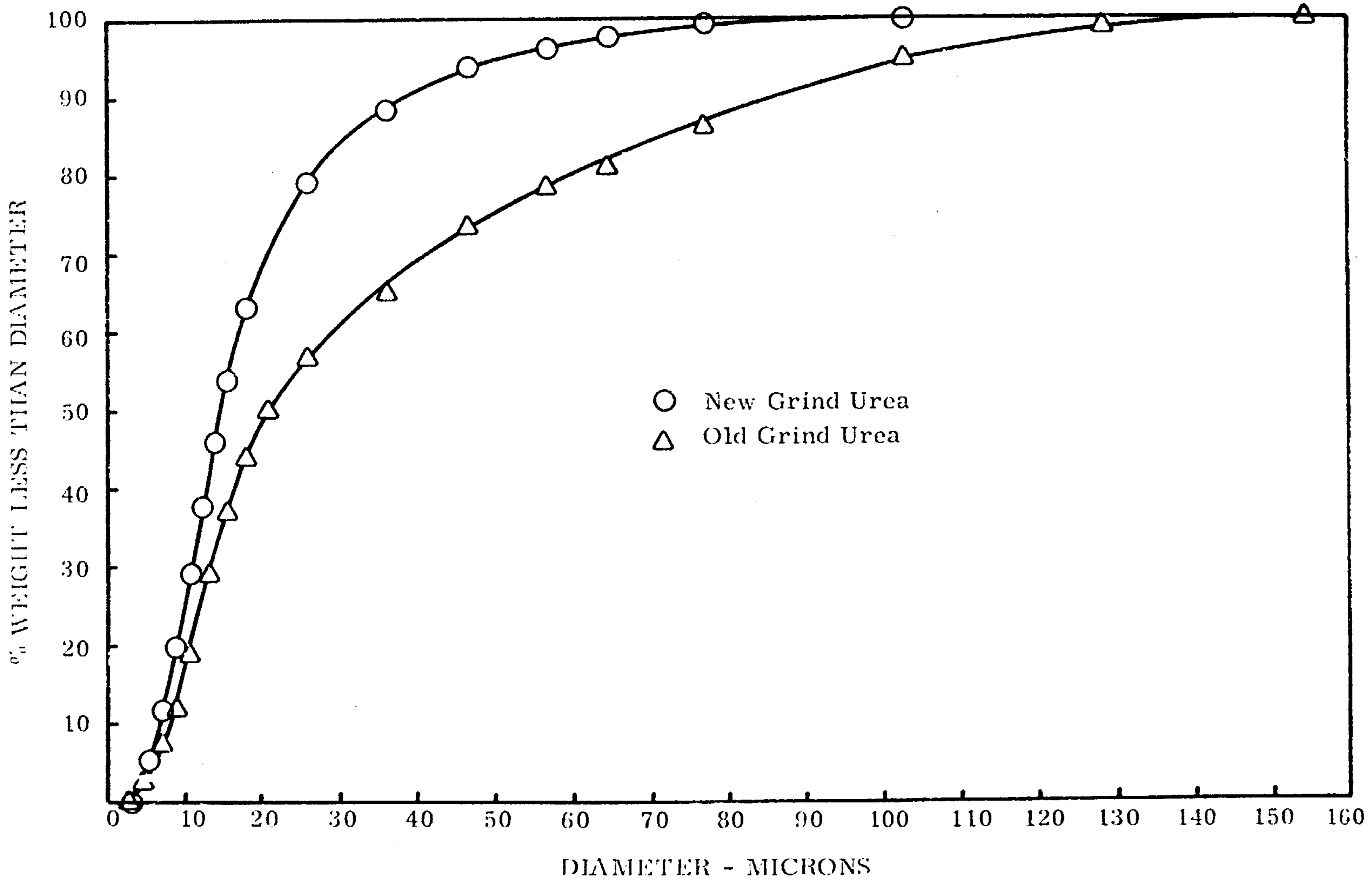
AFCRL desired the technology developed during the sodium chloride studies be applied, if possible, to the relatively innocuous cloud seeding material, urea. Experimental efforts were devoted to this goal, and it was readily ascertained that the ethylcellulose system was amenable, unaltered, to urea encapsulation. Small agglomerates of finely ground urea was effected by encapsulation, and, as with sodium chloride, the encapsulated urea was void essentially of fines.

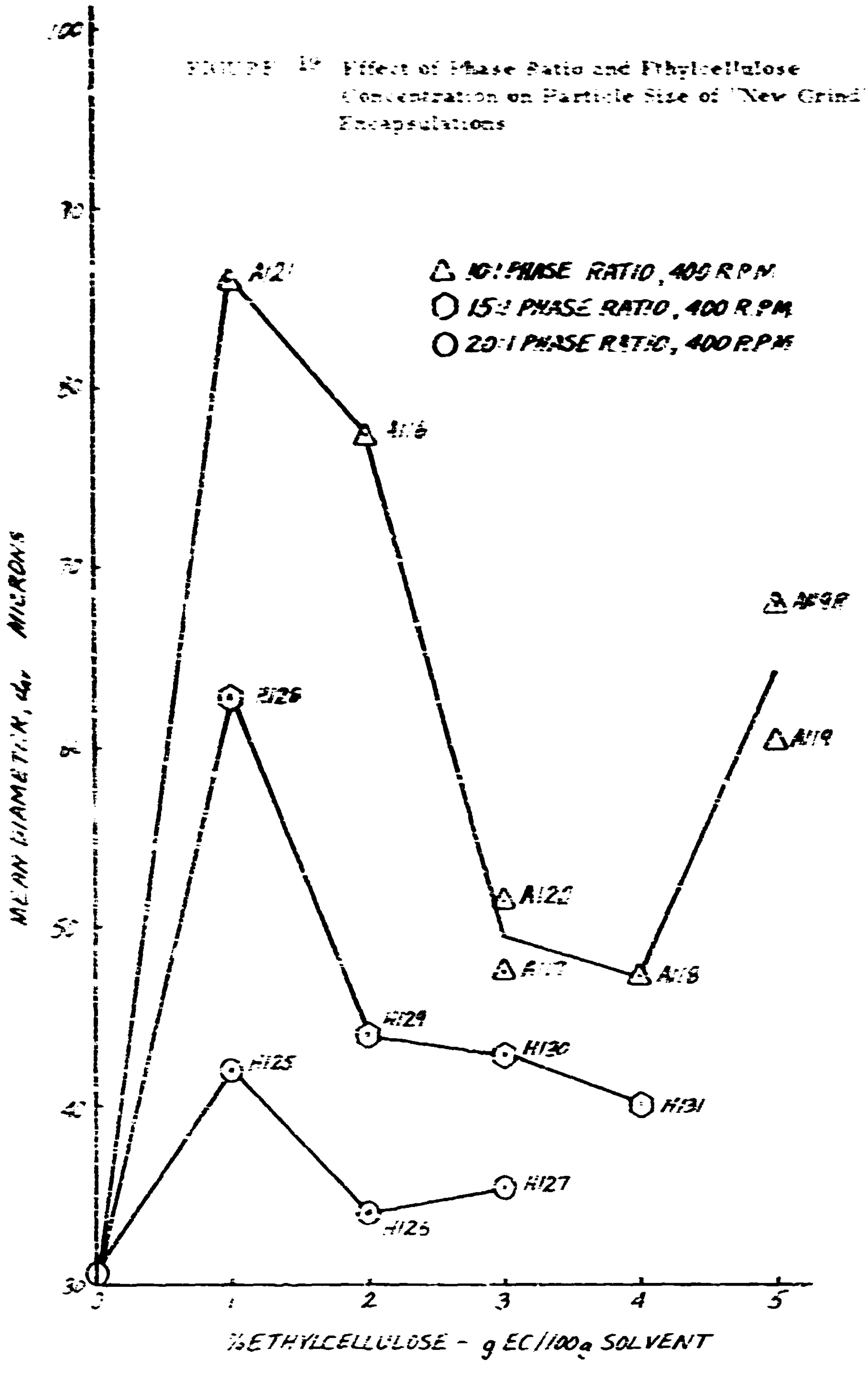
(b) System Optimization and Scale-Up

AFCRL desired that two, diverse particle-size grades of encapsulated urea be available for selection relative to the production of field test quantities. A supplier of finely ground urea, Deepwater Chemical Company, Compton, California, was located with AFCRL's assistance. Two particle-size grinds were received from the supplier for evaluation. The supplier's designation of the materials was "Urea Fog-Dispersant Mix - Old Grind" and "Urea Fog Dispersal Mix - New Grind". The respective particle size distributions, determined by Micromerograph, are given in Figure 18.

Laboratory encapsulation studies were performed to determine the effect of ethylcellulose concentration on the aggregate particle size. Constant agitation was maintained and in the case of the more finely divided raw material (New Grind) encapsulations were prepared at three phase ratios. Namely, 10:1, 15:1 and 20:1 urea: ethylcellulose ratios were employed. The mean diameters, determined by sieve analysis, of the encapsulations are shown versus ethylcellulose concentration in Figure 19. For each phase ratio, the ethylcellulose concentrations employed represent what appeared

FIGURE 18 Particle Size (Weight) Distributions of Urea Raw Materials





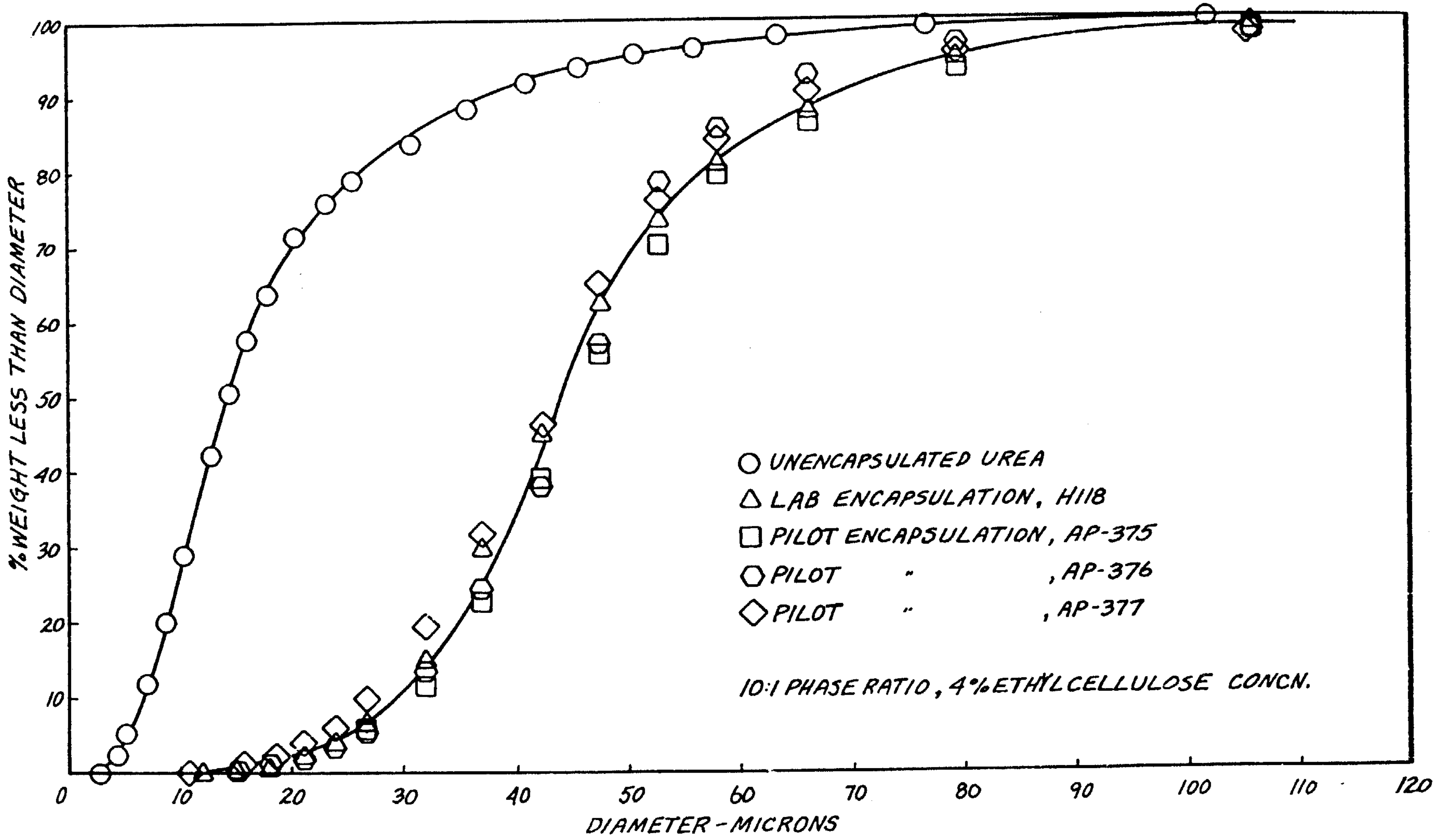
to be the operating range for laboratory and production encapsulation processing. That is, ethylcellulose concentrations in excess to what are shown in Figure 19 did not appear feasible due to the viscosity (total solids, ethylcellulose and urea) of the encapsulation mixture. It is observed that less agglomeration occurs with increasing phase ratios, and that the optimum conditions relative to the particle size objectives of the program appear to be the 10:1 phase ratio prepared at the 3 or 4% ethylcellulose concentration level. Thirty-gallon pilot encapsulations were prepared (Batches AP-375, 376 and 377) at the 4% polymer concentration and the particle size distributions were compared to the laboratory preparation (Sample H-118). The micromerograph size distributions are shown in Figure 20. Also, for comparison the size distribution of the "New Grind" urea, prior to encapsulation, is illustrated.

A similar study was conducted using the more coarse "Old Grind" material. Laboratory encapsulations were limited to a phase ratio of 10:1 and the 4% ethylcellulose concentration appeared optimal with respect to particle size objectives. The results are shown in Figure 21. A single pilot encapsulation was prepared (Batch AP-384) and the particle size distributions of the laboratory and pilot encapsulations are compared to the unencapsulated urea in Figure 22.

Photomicrographs depicting encapsulated aggregates from the pilot encapsulations and their respective urea raw materials are shown in Figure 23. Also, the number distributions for the respective materials are presented as histograms in Figures 24-27.

The results of the pilot encapsulations, compared to the laboratory preparations, were considered excellent and gave assurance of being able to reproduce particle size distributions using production equipment and the urea raw material employed in these tests. As an alternative, it was indicated by laboratory encapsulation

Figure 20 SIZE DISTRIBUTION OF LABORATORY AND PILOT PLANT ENCAPSULATIONS, SMALL GRADE UREA



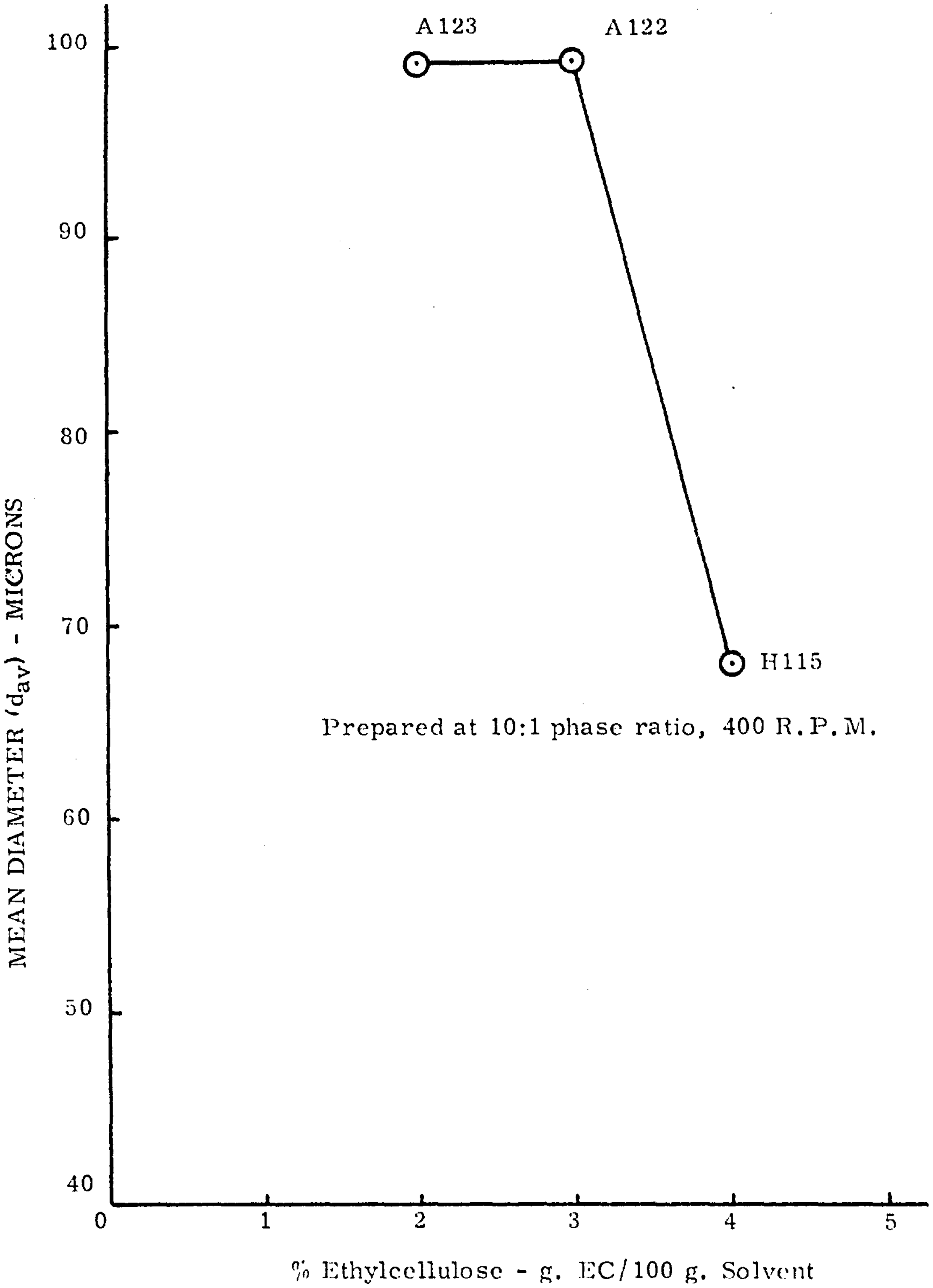


FIGURE 21 Effect of Ethylcellulose Concentration on "Old Grind" Encapsulations

FIGURE 22 Size Distribution of Laboratory and Pilot Plant Encapsulations Large Grade Urea

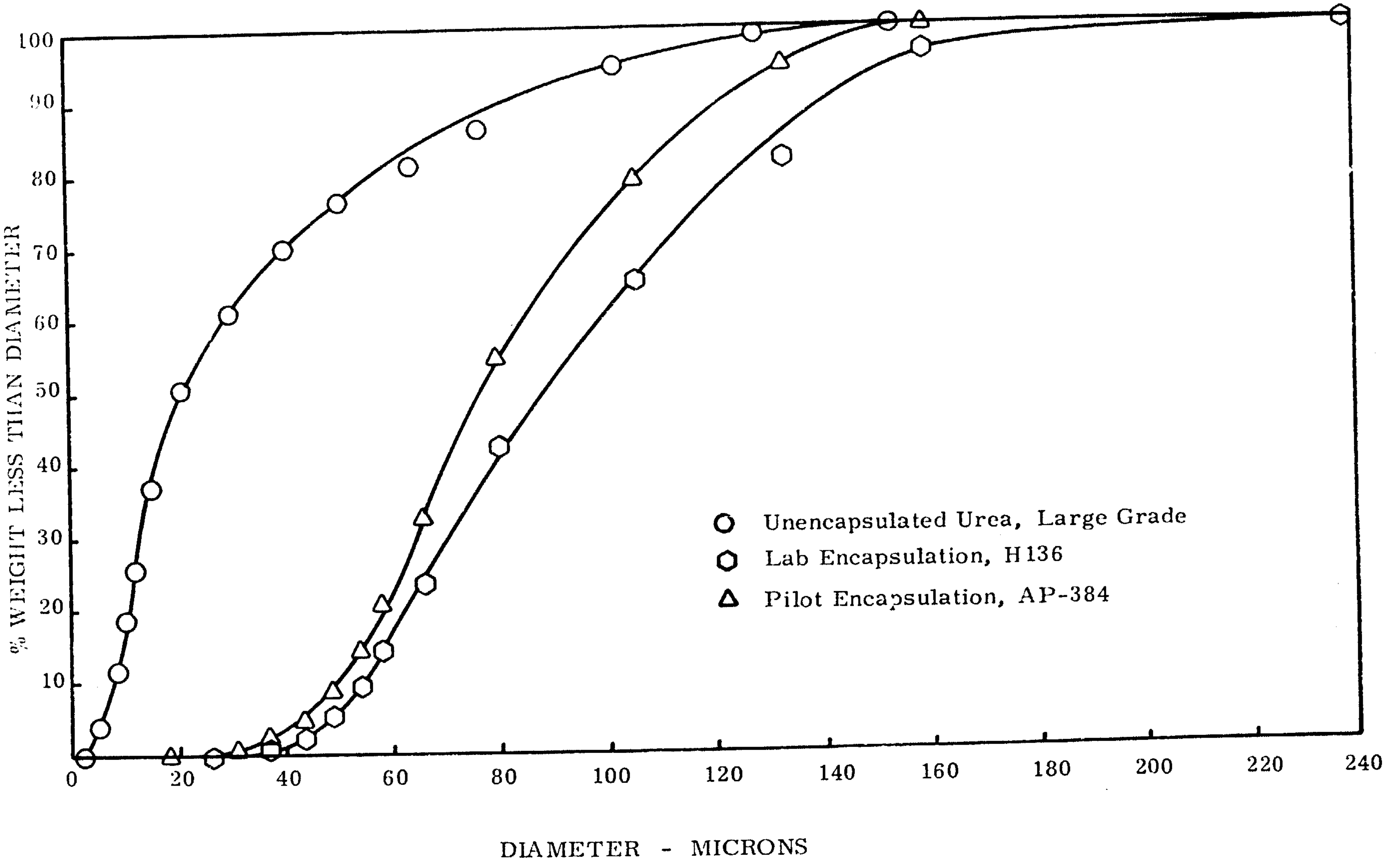
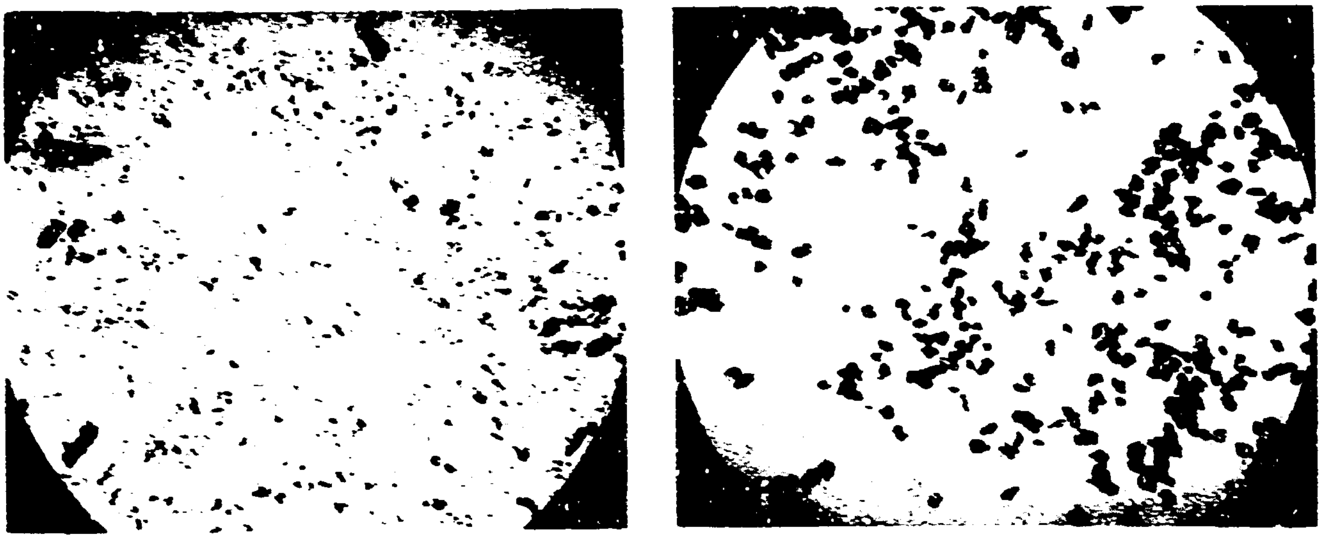


FIGURE 23

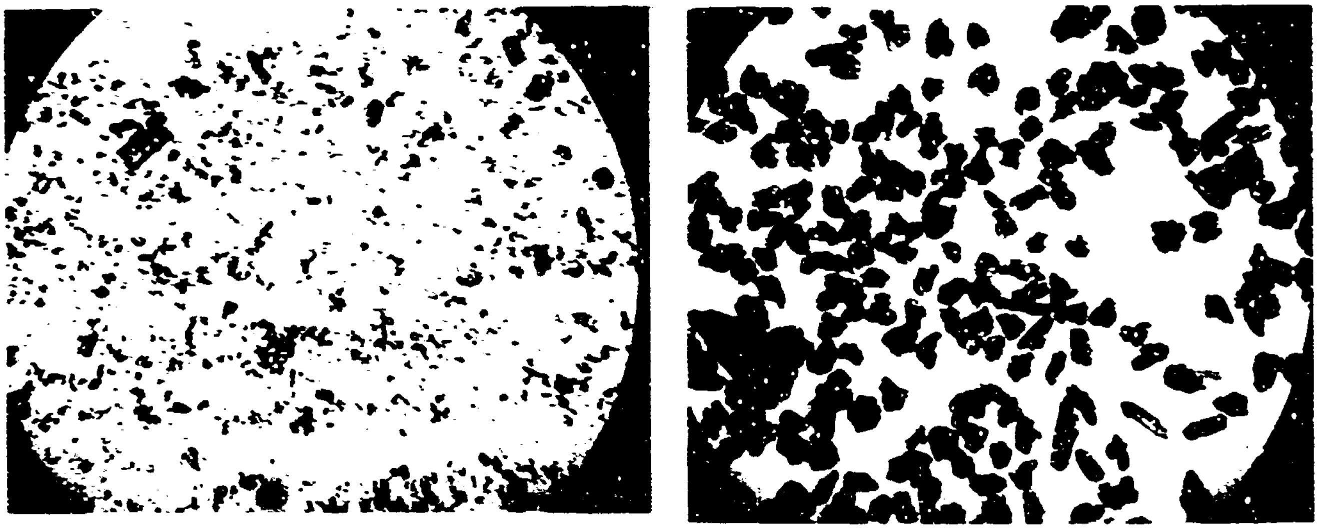
PHOTOMICROGRAPHS OF UREA GRADES BEFORE AND AFTER PILOT ENCAPSULATIONS



Small Grade Before Encapsulation

Small Grade After Encapsulation

Large Reticle Square = 70 Microns



Large Grade Before Encapsulation

Large Grade After Encapsulation

Large Reticle Square = 70 Microns

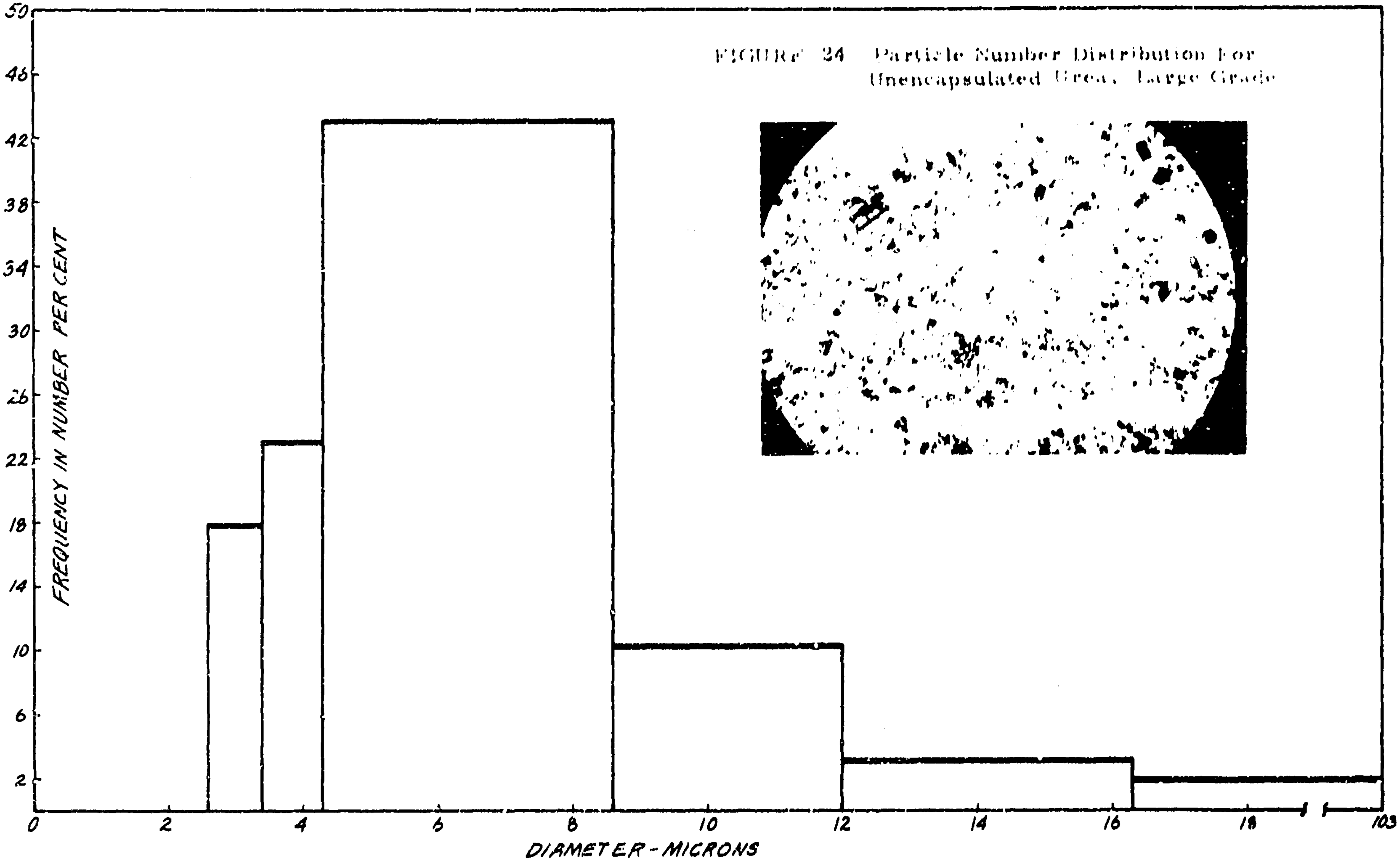


FIGURE 24 Particle Number Distribution For Unencapsulated Urea, Large Grade

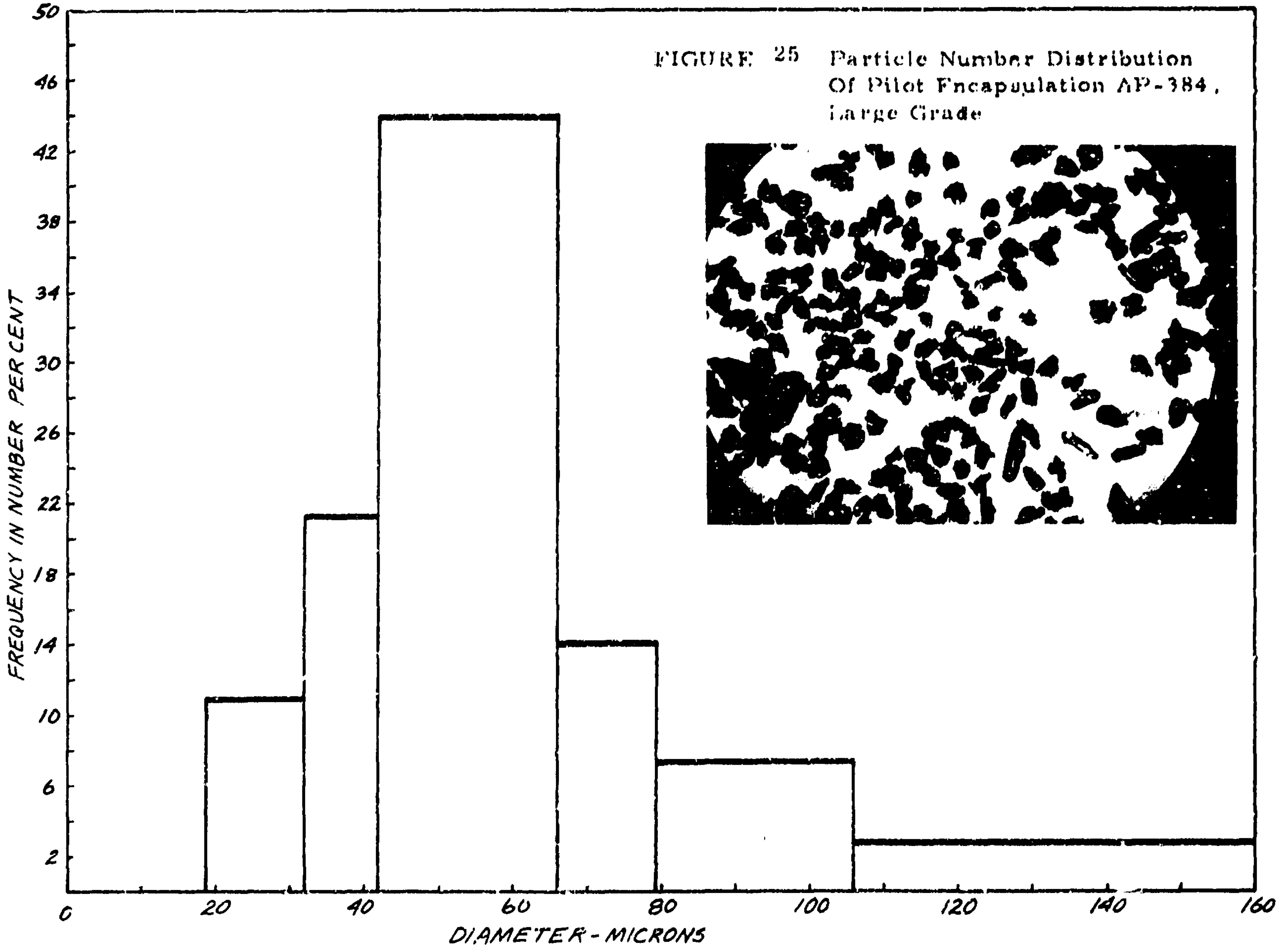


FIGURE 25 Particle Number Distribution Of Pilot Encapsulation AP-384, Large Grade

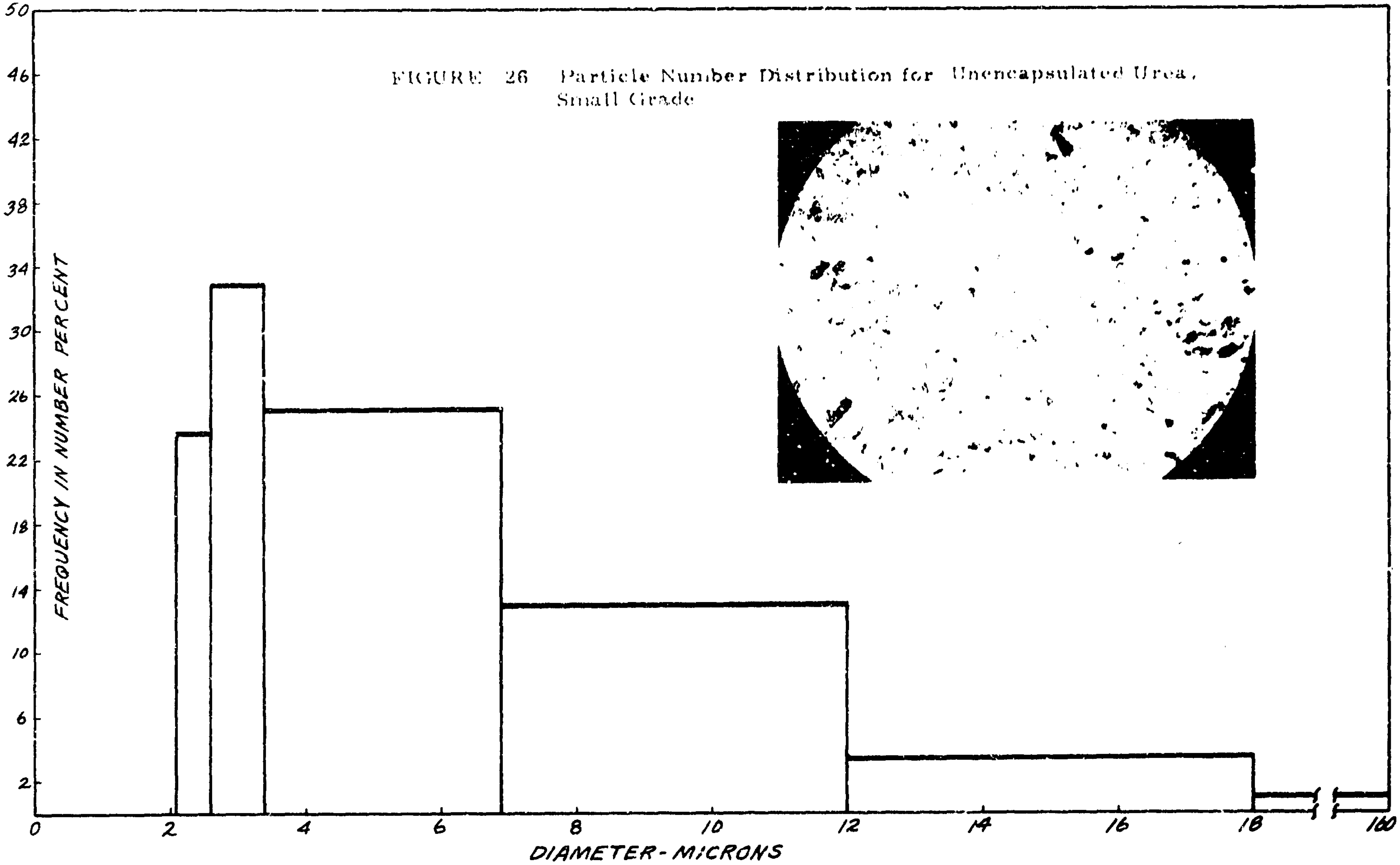


FIGURE 26 Particle Number Distribution for Unencapsulated Urea, Small Grade

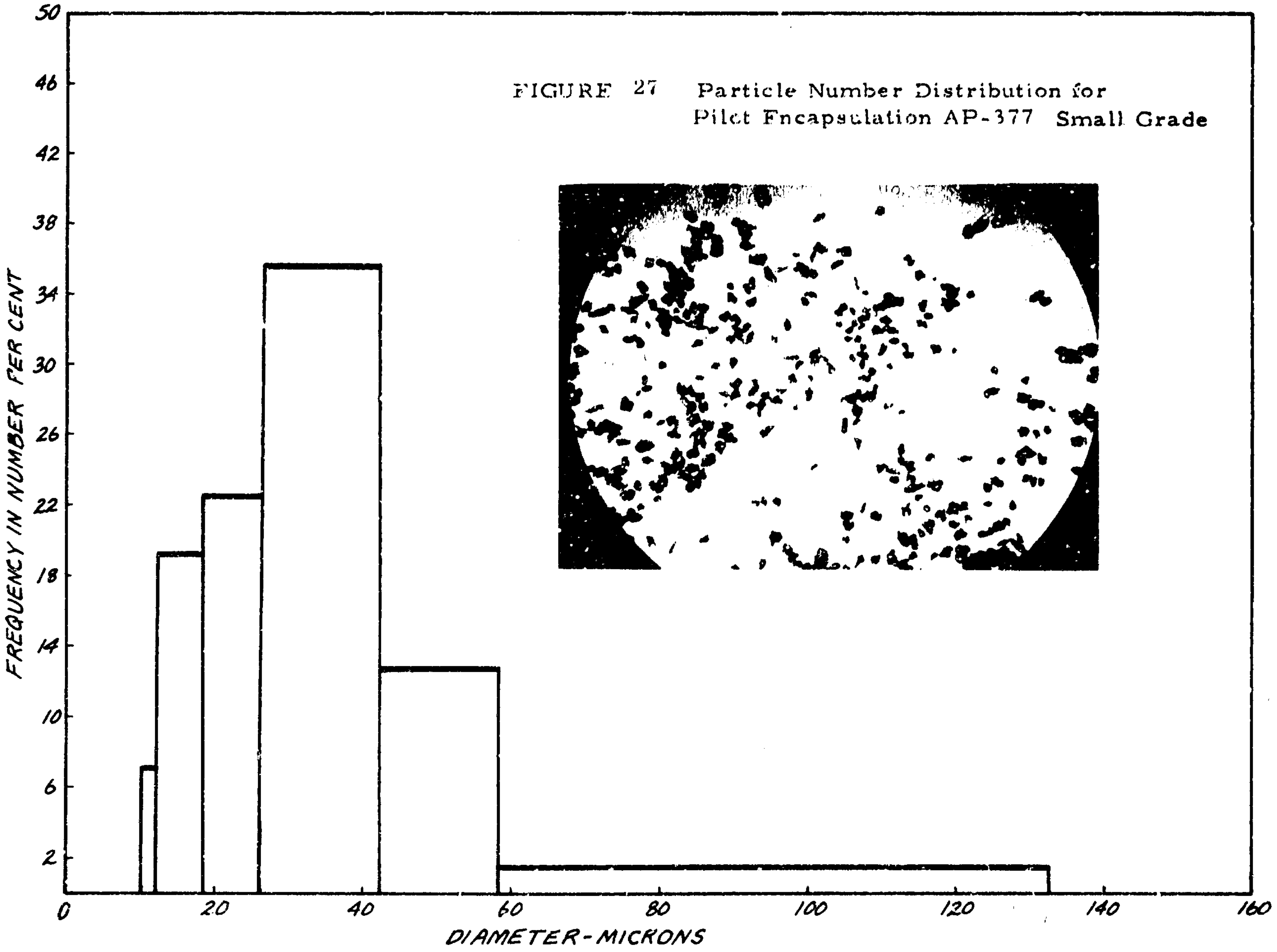


FIGURE 27 Particle Number Distribution for Pilot Encapsulation AP-377 Small Grade

that another processing variable, agitation, could be employed to produce larger aggregates, if desired, using the more finely divided (New Grind) raw material. The size distributions of laboratory encapsulations prepared at diverse agitation rates are shown in Figure 28.

The results of the study revealed that by encapsulating finely divided urea with ethylcellulose, aggregates of varied dimensions (diameters) could be produced. Processing variables affecting the particle size distribution and mean diameter were size distribution of the urea raw material, phase ratio (urea to ethylcellulose), ethylcellulose concentration of encapsulation mixture and rate of agitation.

2. Water Sorption Properties

The optical method described previously was utilized to ascertain the water sorption properties of the urea particles. The results are shown graphically in Figure 29. Each point on the curve represents the average value for at least five particles. The data indicated that the encapsulated aggregates exhibited water sorption properties comparable to those obtained with unencapsulated urea particles of similar dimension. Figure 30 illustrates photomicrographs of the condensation occurring when the particles were exposed to the humid (ca 100% RH) environment.

Also, the water sorptive properties of encapsulated urea aggregates were verified by AFCRL with cloud chamber tests conducted at Cornell Aeronautical Laboratory, Inc.

3. Bulk Powder Properties

The caking tendencies of unencapsulated and encapsulated urea bulk powders were determined by the test method previously described. As was found with the sodium chloride studies, the ethylcellulose-encapsulation of urea grossly reduced the caking tendencies of the bulk powder when exposed to a humid environment (32°C, 87-93% RH) for 120 minutes. All encapsulations prepared with the smaller

FIGURE 28 Effect of Agitation on Particle Size of "New Grind" Encapsulations

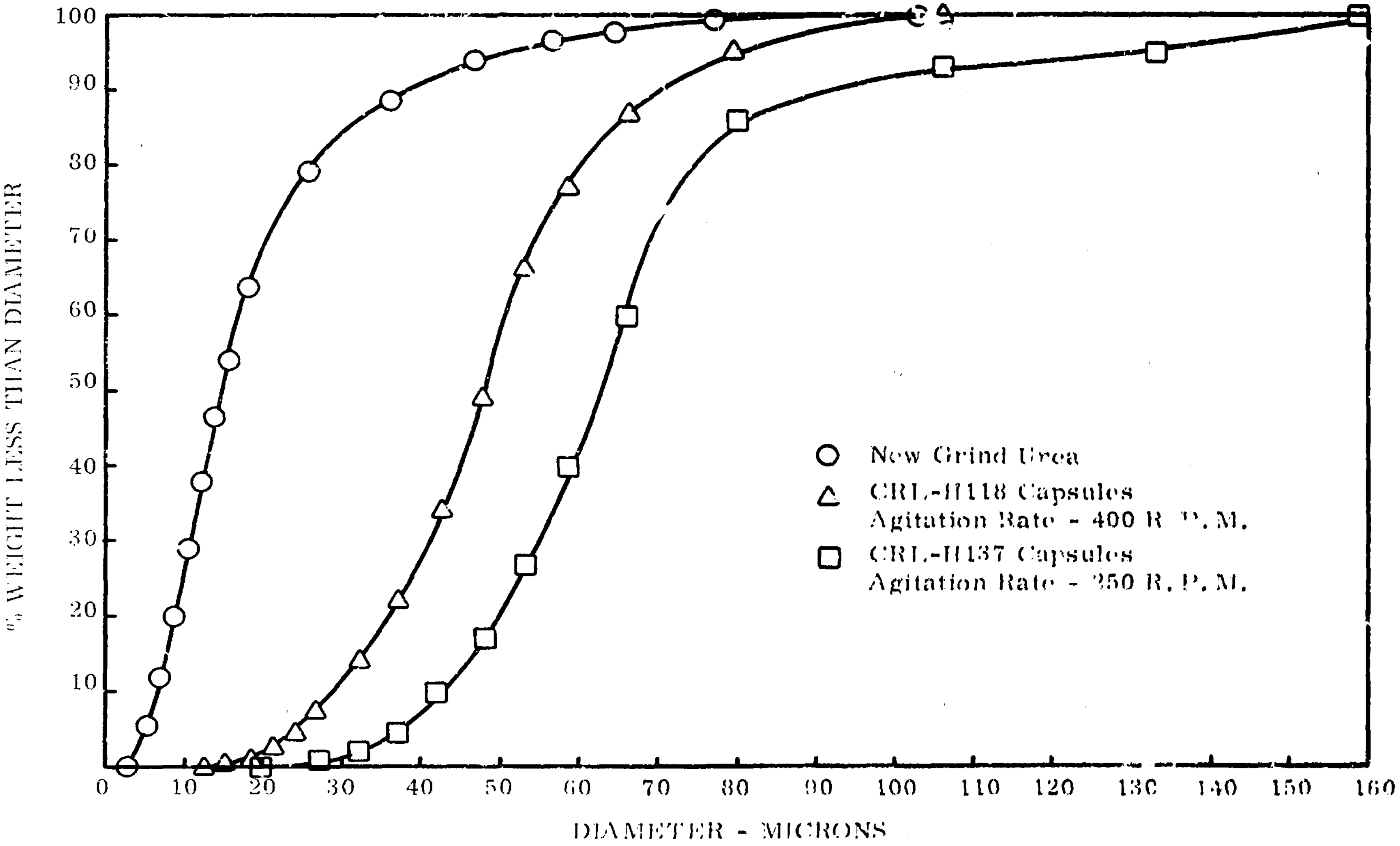


FIGURE 29

Growth of Urea Particles at 100% RH, 28°C

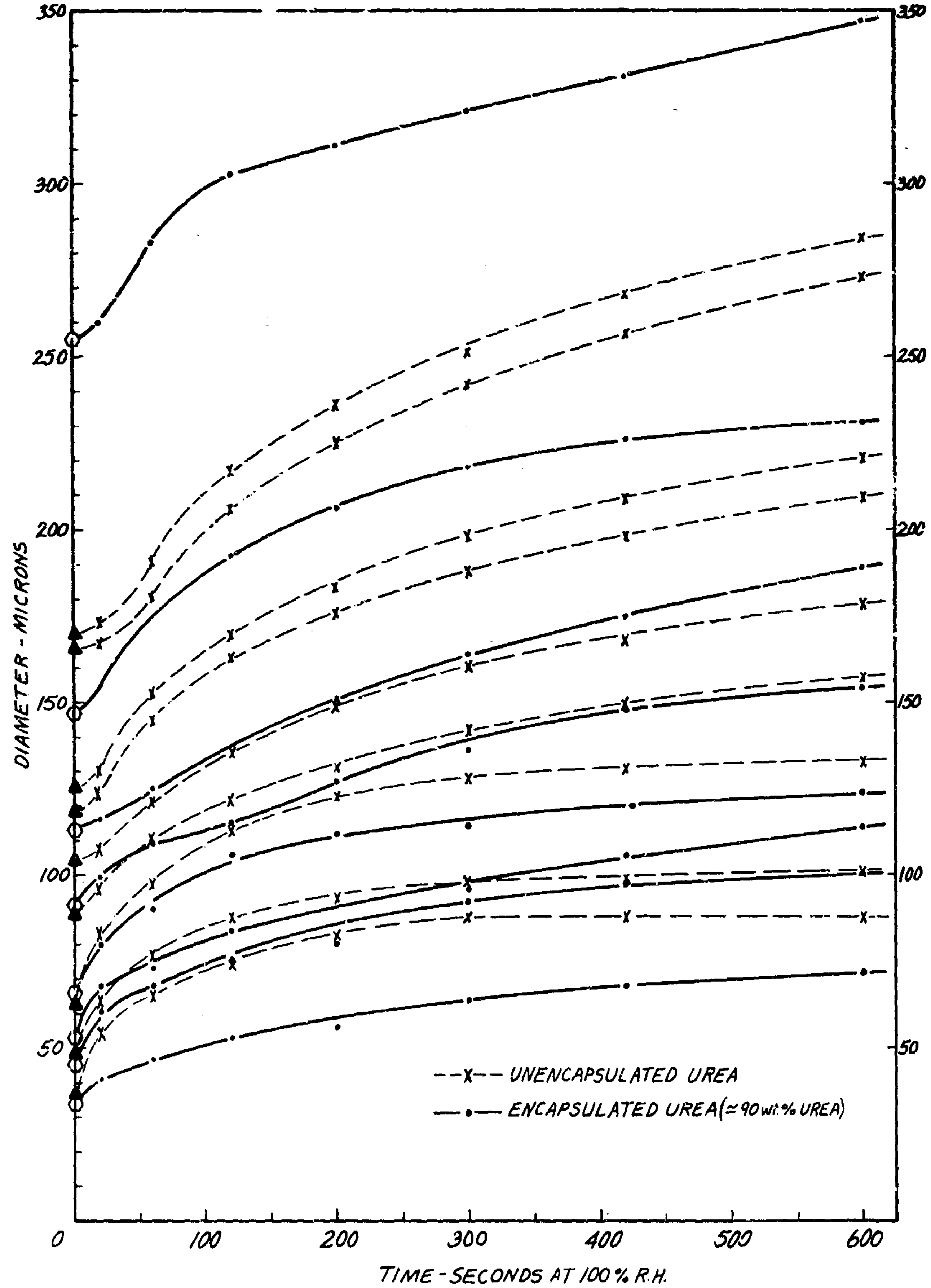
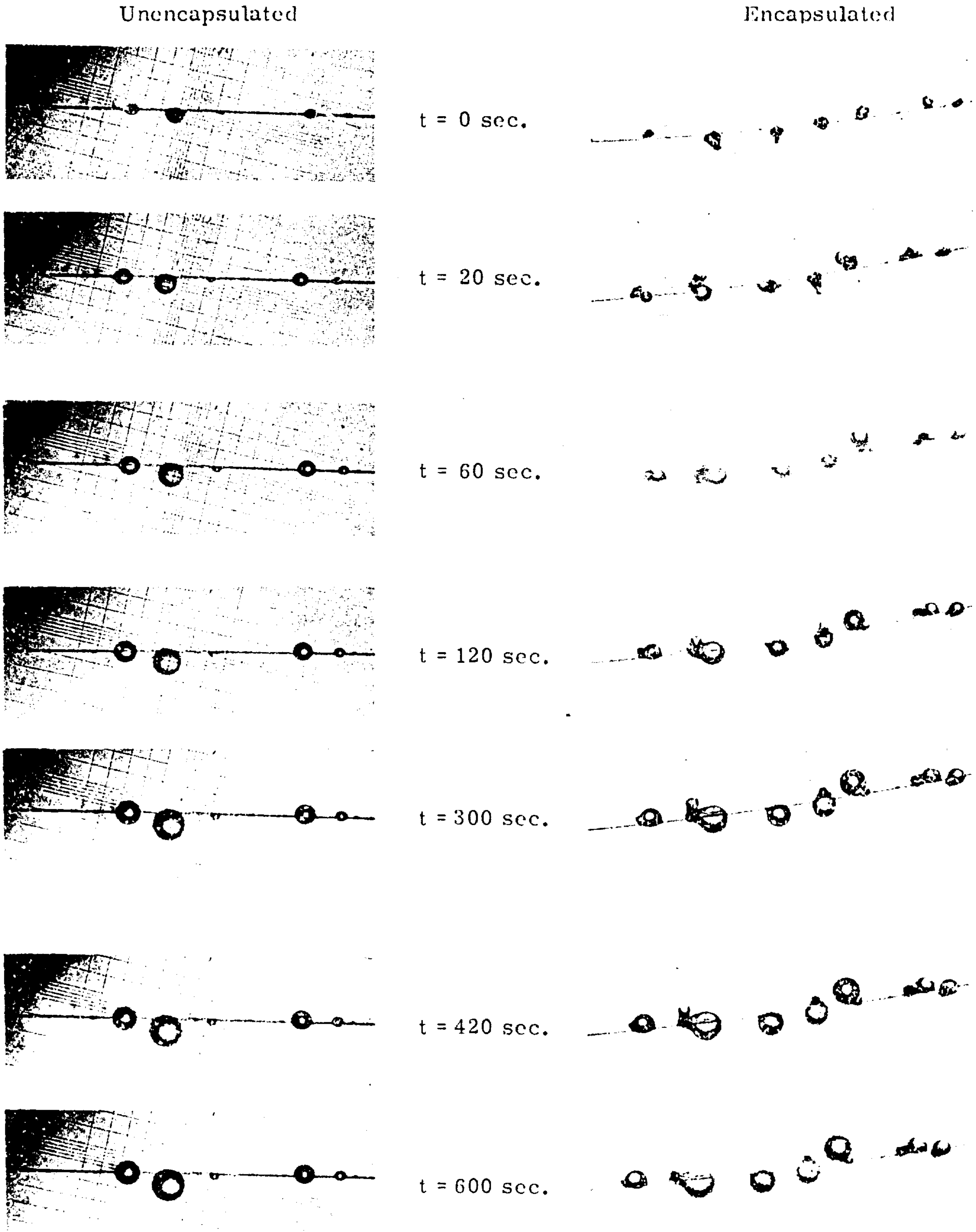


FIGURE 30

Growth of Urea Particles at 100% RH, 28°C

(Large Reticle Square = 70 Microns)



"New Grind" material demonstrated equivalent reduction of agglomeration irrespective of the phase ratio or particle size of the encapsulation. Encapsulation of the more coarse "Old Grind" also substantially decreased the agglomerating tendency of the bulk powder. The results are tabulated below.

Bulk Powder Agglomeration

	% Retained On 100 Mesh Screen Before Exposure	% Retained On 100 Mesh Screen After Exposure
Unencapsulated "New Grind"	0%	70-80%
Encapsulated "New Grind"	0%	2-8%
Unencapsulated "Old Grind"	5%	80-90%
Encapsulated "Old Grind"	7%	10-16%

The agglomerating tendencies of the powders are compared in the photographs presented in Figure 31.

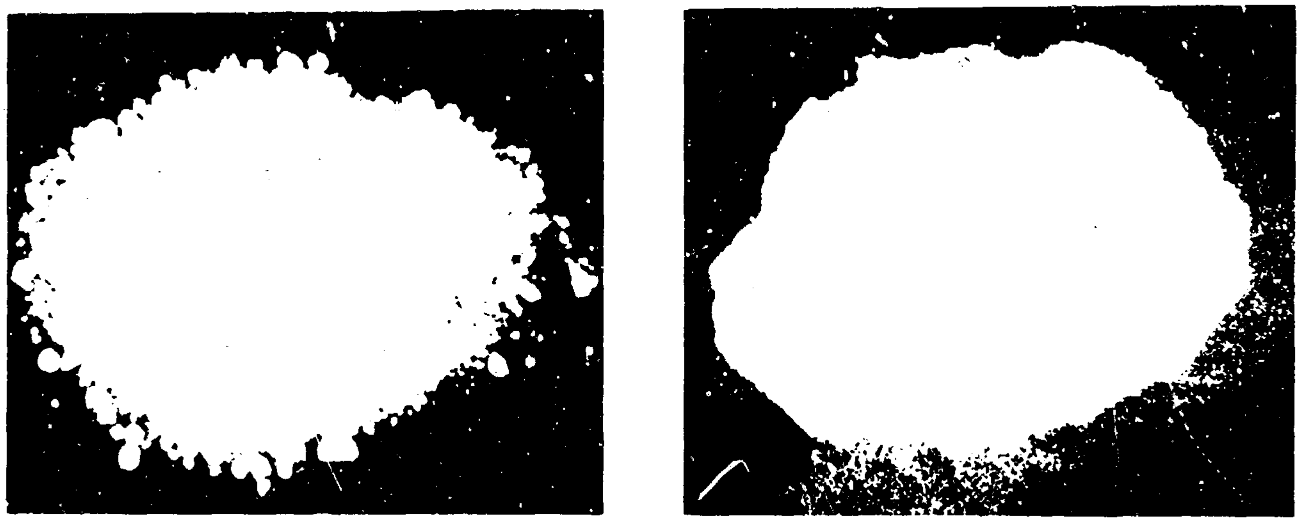
The amount of moisture sorbed by the bulk powders during the 120 minute exposure to the 32°C, 87-93% RH environment was determined quantitatively. The results showed the encapsulated and unencapsulated urea sorbing the same amount of moisture. Within the test conditions, the bulk powders sorbed from 1.0 and 1.2% moisture. This data indicated that the reduction of the agglomerating tendency effected by ethylcellulose encapsulation is related to surface and interfacial phenomena rather than to any reduction of the amount of moisture sorbed. It should be noted that the moisture sorption and agglomerating characteristics of the bulk powders will be related to the exposed surface - total volume ratio of the powder sample. In this particular test the ratio is 13 $cm^2/50\ cm^3$.

FIGURE 31

Urea Bulk Powders After Open-Container Exposure
To 32°C/90% RH for 120 Minutes

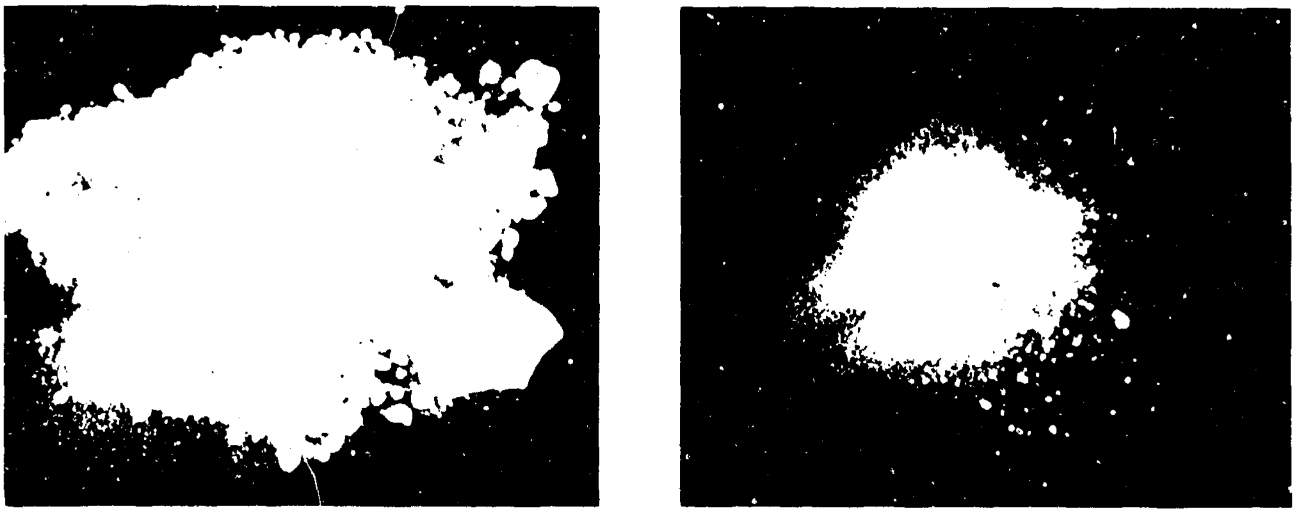
Unencapsulated Powder

Encapsulated Powder



Total Sample

Total Sample



Fraction Retained on
100 Mesh Screen (80 wt. %)

Fraction Retained on
100 Mesh Screen (80 wt. %)

D. PRODUCTION OF ENCAPSULATED UREA

1. Selection of Materials

The results of the developmental efforts involving the ethylcellulose encapsulation of sodium chloride and urea, allowed AFCRL to make a selection from three candidate production materials. The characteristic properties of the three materials, two grades of encapsulated urea and a single grade of sodium chloride, are given in Table V. AFCRL advised that the field test quantities would be limited to hygroscopic agent, urea. Also, the production of the initial 6,000 pounds was to be comprised of 3,000 pounds of the Small Grade and 3,000 pounds of the Large Grade urea. Selection of the particular material for the remaining 14,000 pound production was to be made by AFCRL pending the results of their numerical model studies.

2. Production

Production of the Large Grade material was commenced in late July, 1970, using a 200-gallon production facility. An output of about 350 lbs./batch was realized in the batch process. Each batch was analyzed with respect to particle size and particle distribution using the Sharples Micromerograph. The finished product was packaged in polyethylene bottles and cardboard shipping containers. Each container was filled to 25 ± 0.5 lbs. The cartons were individually labelled according to encapsulation batch number and pallet identifying number. The finished cartons were palletized; 27 cartons (675 lbs.) comprising a shipping pallet.

The production equipment and processing steps are pictorially presented in Appendix I. Also, found in Appendix II are the number distributions and histograms of production batches. The shipping pallet composition, by encapsulation batch number and mean diameter, are given in Appendix III for all materials delivered to AFCRL for field tests.

TABLE V

Encapsulated Materials for Production Selection

Cloud Seeding Material	Encapsulated Sodium Chloride	Encapsulated Urea Large Grade	Encapsulated Urea Small Grade
Active Content	90 wt. % Sodium Chloride	90 wt. % Urea	90 wt. % Urea
Coating Material	Ethylcellulose	Ethylcellulose	Ethylcellulose
True Density	2.000 g/cm^3	1.275 g/cm^3	1.275 g/cm^3
Bulk Density	0.59 g/cm^3	0.53 g/cm^3	0.51 g/cm^3
Particle Size Distribution	30-80 microns (90% of material by wt.)	40-140 microns (90% of material by wt.)	20-80 microns (90% of material by wt.)
Mean Diameter, d_{av}	62 microns	82 microns	45 microns
Water Vapor Sorption of Dispersed Particles at 100% RH	Comparable to unencapsulated NaCl particles of equivalent dimensions	Comparable to unencapsulated Urea particles of equivalent dimensions	Comparable to unencapsulated Urea particles of equivalent dimensions
Bulk Powder Agglomeration at 120 minutes at 32°C/90% RH*	2% of sample, by wt., greater than 149 microns	12% of sample by wt., greater than 149 microns**	6% of sample, by wt., greater than 149 microns
Developmental Reference Material	Sample CRL-A102	Sample AP-384	Sample AP-377

* Exposed surface to total volume ratio = 13 cm^2/50 cm^3

** 5% of material is greater than 149 microns before exposure

Production of the Large Grade material, batches AP 432-434, UC 4-11, was completed and the Small Grade production was commenced. An equipment limitation was encountered during the initial production of the Small Grade (Samples UF 1-6, Appendix II). The capacity of a solvent dryer was exceeded with the smaller particles causing additional labor costs to be incurred. AFCRL was apprised of the problem, and it was decided to devote the remainder of the program to the production of Large Grade material. The decision was based on AFCRL's numerical model results indicating the Large Grade to be more amenable to the fog-clearing problem.

Production of the Large Grade urea was resumed, and after approximately 13,000 pounds of material had been produced, a potential problem was reported. A 7,000 pound shipment, transported from Dayton, Ohio, to Altadena, California, via motor freight in September, 1970, (see Appendix III) was sampled by AFCRL's contractor, Meteorology Research, Inc. (MRI). The contractor found indications that the encapsulated urea shipment was undergoing clumping and caking. The degree of clumping, although not appearing excessive at the time of sampling, was believed to be time dependent based on MRI's experience with other hygroscopic fog seeding agents. It was the opinion that additional aging would constitute a serious problem regarding the efficiency of the material at the time of use. A NCR technical representative traveled to MRI to observe and discuss the potential problem with MRI personnel. Sampling the material showed two characteristics:

(1) The presence of small, hard particles which were a few thousand microns in size, and could not be broken down, by hand, to their fundamental microcapsule size.

(2) The presence of relatively larger amounts of large, soft clumps which were broken down by slight physical action.

With respect to (1), samples of the batches retained by NCR were subjected to a sieve analysis and it was found that an average value, for the 23 batches in question, of 6%, by weight, would not pass a 100 mesh (149 micron) screen. Micromerograph analyses at the time of production showed an average of 3%, by weight, being greater than 133 microns in diameter. The small clumps were the result of residue accumulating in the solvent dryer. Also, at the time of packaging and analysis, the clumps were soft and friable. Consequently, the deagglomerating mechanism of the micromerograph particle-size analyzer reduced the small clumps, in the main, to their fundamental capsule particles. Although these small clumps were not the main concern as they represented less than 0.5% of the total particles, it was decided to add a screening or sieving step to the process prior to packaging.

Regarding the potentially serious problem presented by (2) above, the laboratory testing of developmental samples of encapsulated urea indicated that gross clumping would not occur. The tests, however, did not involve the pressures and compaction which could occur on transporting and storing the final, packaged products. Being unable to extrapolate from the laboratory data with absolute confidence, it was decided to add an anticaking agent to the encapsulated materials. MRI suggested that tricalcium phosphate (TCP) be added to the encapsulated urea at a level of 2 weight percent. Consequently, it was decided that MRI would dry blend TCP with the existing 13,000 pounds of material produced.

A small portion of the material, sufficient in quantity to conduct dissemination tests, was not blended with TCP for subsequently comparing the effects of the anticaking additive.

The production of the remaining Large Grade urea, about 7,000 pounds, included the addition of TCP. The anticaking agent was added to the encapsulation slurry and dispersed with agitation prior to the normal filtration and solvent removal processing steps. Final delivery of the encapsulated materials was made to AFCRL in December, 1970 (see Appendix III).

The approximate composition of the encapsulated materials delivered is given below.

Skids No. 1-20 and No. 31 (See Appendix III)

Ethylcellulose	9.1% by wt.
Urea	87.9% by wt.
Grinding Dispersant	1.0
Biuret	1.8
Cyanuric Acid	0.2

Skids No. 21-30 (See Appendix III)

Ethylcellulose	9.0% by wt.
Urea	86.5
Grinding Dispersant	1.0
Biuret	1.8
Cyanuric Acid	0.2
Tricalcium Phosphate	1.6

Limited sampling of packaged materials showed moisture contents of 0.5% or less.

The limited production run (20,000 lbs.) of encapsulated urea made obvious the inability to maintain batch-to-batch replication of particle size and distribution. The process, in general, produced deviations greater than those expected. In addition, the encapsulation process was found less responsive to adjustments in agitation, ethylcellulose concentration and phase ratio than was observed during the development studies. The average cumulative particle distribution of 58 production batches is given in Figure 32. Also, presented in the figure are the mean deviation, d, and the range of the production batches.

Elucidation of the problem regarding particle size control was not achieved until late in the production run. The degree of replication expected of the process was achieved finally by discarding the practice of reusing the solvent media, cyclohexane. Essential to the economy of the encapsulation process is the ability to continue to use the solvent, following filtration of the encapsulated material, for subsequent encapsulation. With most materials, this practice has been feasible without resorting to purification of the filtrate (solvent) via distillation, prior to reuse. With encapsulated urea processing, however, it was found that optimal particle size control could be achieved and maintained only by using fresh solvent media for each encapsulation batch. This factor does not reflect substantial increases in encapsulation costs for large scale production; however, in the production of quantities such as was achieved in this program, this does present added processing costs. Charging the system with fresh cyclohexane improved the batch-to-batch replication of particle size and distribution. Also, the process became sensitive to variation of certain parameters which were found influential to particle size control during the developmental efforts.

The results of the production study have allowed the specification listed below to be considered attainable regarding the replication of encapsulation batches.

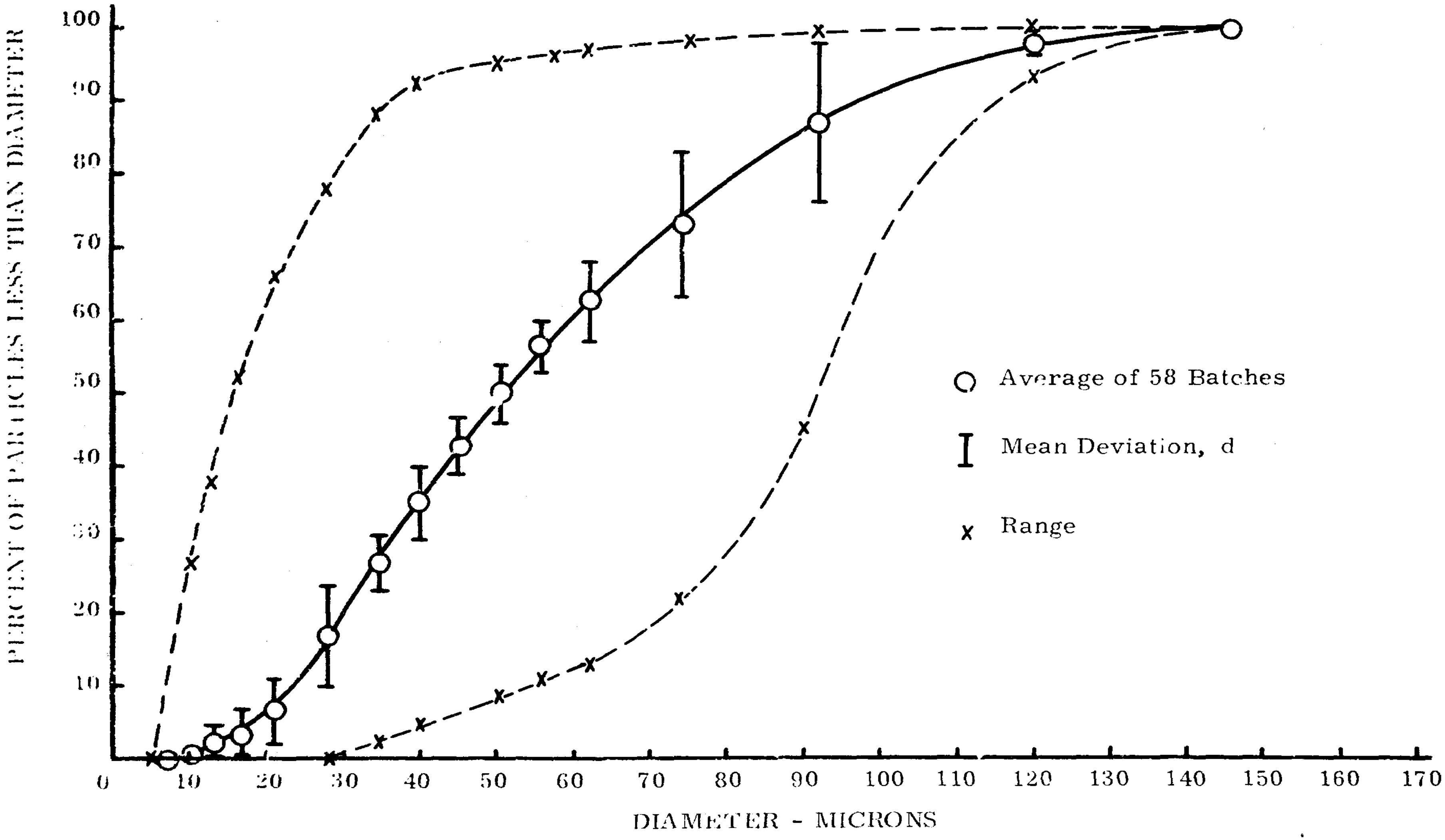
Median particle number diameter	= 50 ± 20 microns
Percent of particles 10-80 microns in diameter	= 85% minimum
Percent of particles less than 10 microns in diameter	= 1% maximum

3. Warm Fog Dispersal Trials

AFCRL's warm fog dispersal testing of encapsulated urea were to be conducted to McClellan AFB, Sacramento, California, during January-February, 1971. During the week of January 11, an NCR technical representative participated in those tests. Unfortunately, during the period of NCR participation, appropriate fog test conditions did not avail. However, the field test operations, equipment, facilities design, etc. was presented and discussed by the contributing personnel of AFCRL, its contractor, MRI, and McClellan AFB. During the discussions, it was reported that helicopter dissemination tests had shown that the encapsulated urea containing no anticaking additive, TCP, exhibited no additional clumping or caking problems when compared to the encapsulated materials containing the additive.

Telephonic reports received from AFCRL personnel regarding fog dispersal tests conducted subsequent to the NCR visitation have indicated that the encapsulated urea has been found to be functional. The material exhibited improved handling and dissemination properties and behaved hygroscopically when aerially dispersed in warm fogs.

FIGURE 32 Particle Distribution of Large Grade Urea Production Batches



SECTION III

CONCLUSIONS

An effective process has been developed for encapsulating with ethylcellulose the hygroscopic cloud seeding materials, urea and sodium chloride. The process is applicable to other materials which meet the following criteria:

The materials to be encapsulated must

(1) be a finely-divided powder,
(2) be insoluble in cyclohexane, and
(3) must be compatible and non-reactive to ethylcellulose and cyclohexane.

The encapsulation process effects agglomeration of the finely divided powder. The degree or amount of aggregation occurring during encapsulation is related to the following processing parameters:

(1) The particle size and distribution of the material to be encapsulated.
(2) The agitation applied during the batch-encapsulation process.
(3) The polymer, concentration of the coating solution.
(4) The amount of coating polymer applied onto the powder particles.

The above parameters can be employed to vary and control the particle size and distribution of the encapsulated aggregates; however, the particle size range, relative to minimum and maximum limits achievable, were not ascertained during the program. The capability was developed for achieving encapsulated urea in production quantities having the following particle size properties.

Median Particle number diameter = 50 ± 20 Microns

Percent of particles 10-80 microns in diameter = 85%, minimum

Percent of particles less than 10 microns in diameter = 1%, maximum

A unique property resulting from encapsulation processing is that the process yields an encapsulated product which is void, essentially, of fines--an important aspect relative to effective warm fog seeding agents. In addition, the encapsulated hygroscopic powders demonstrate resistance to clumping and caking during normal storage and handling procedures. The anticaking behavior is related to surface and interfacial phenomena and not to the reduction of the hygroscopicity of the bulk powders.

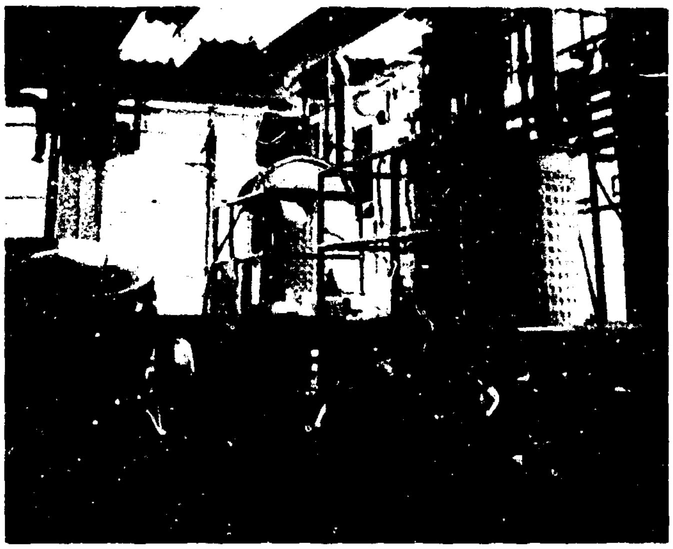
Water sorption and particle growth through condensation is accomplished by diffusion mechanisms operating across the permeable, water insoluble coating polymer, ethylcellulose. Laboratory tests reveal the encapsulated aggregates to be comparable in hygroscopicity to unencapsulated particles having equivalent dimension. The intraparticle porosity or internal surface area of the encapsulated particles is believed to be instrumental to the relatively uninhibited water sorption properties.

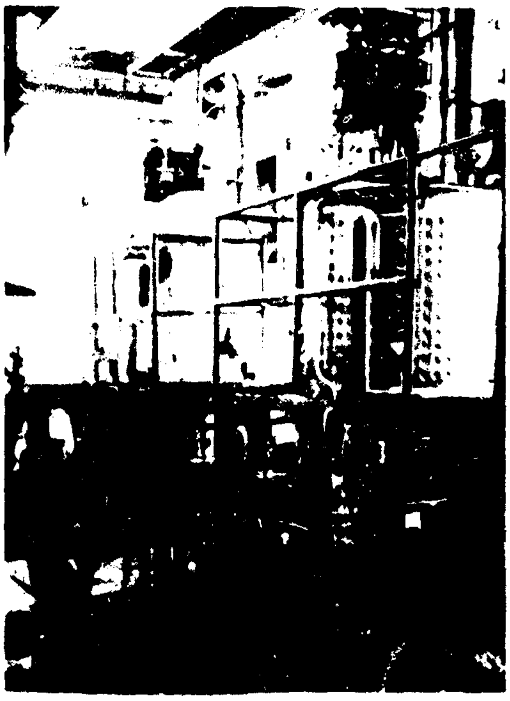
The effectiveness of the encapsulated materials, relative to their water sorption properties, was demonstrated in laboratory warm fog atmospheres independently by the laboratory tests of NCR and AFCRL and by AFCRL warm fog dispersal field tests.

APPENDIX I

PICTORIAL SEQUENCE OF ENCAPSULATED UREA PRODUCTION

PRODUCTION SEQUENCE

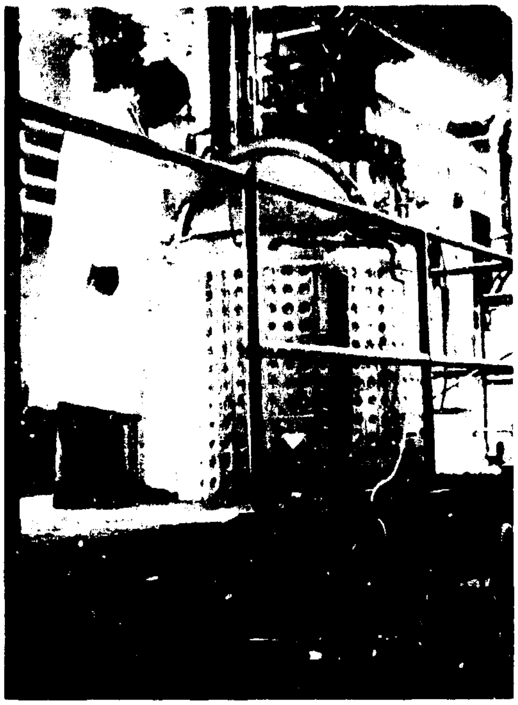




200-Gallon Encapsulation Facilities
keted, Agitator Equipped Encapsulation Vessels

PRODUCTION SEQUENCE CONTINUED

Charging Vessel with Urea

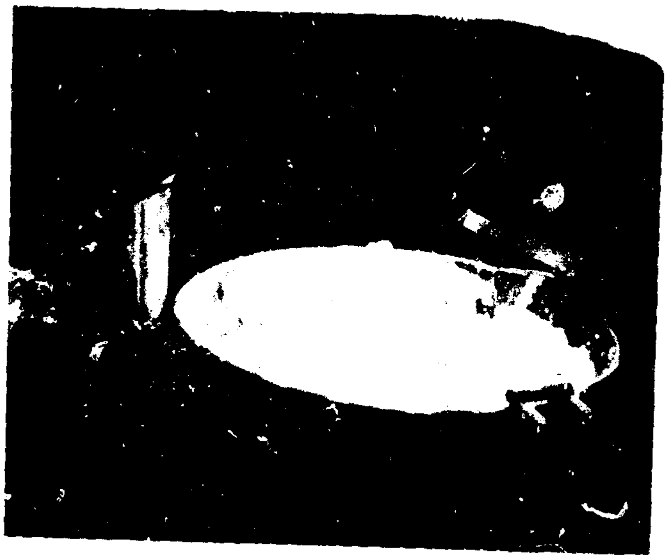


Operator Inspecting Dispersion

Filtration Apparatus

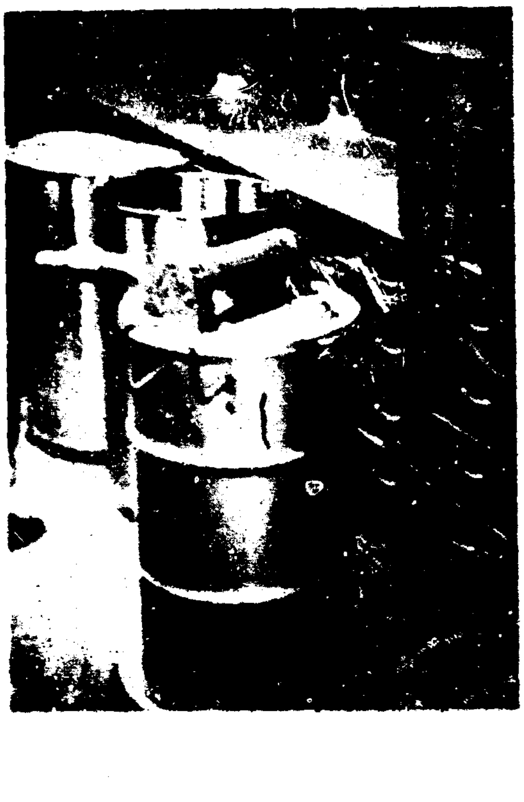
Solvent Dryer

PRODUCTION SEQUENCE CONTINUED



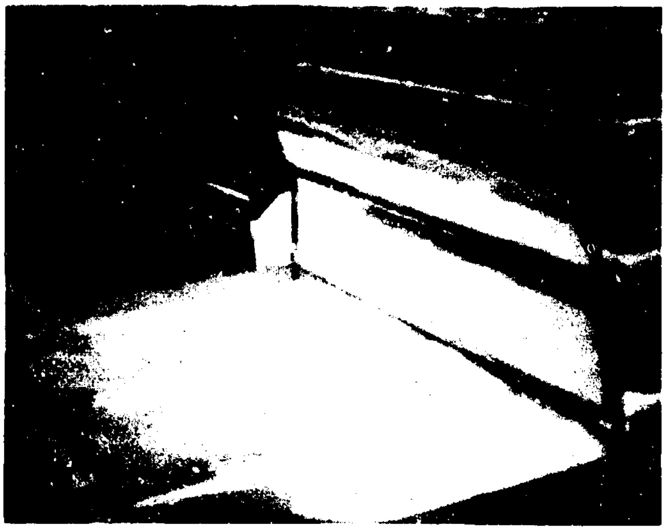
Product After Filtering

Loading Dryer



Vacuum Unit for Loading Bottle-Filling Apparatus

PRODUCTION SEQUENCE CONTINUED



Unloading Finished Product from Solvent Dryer

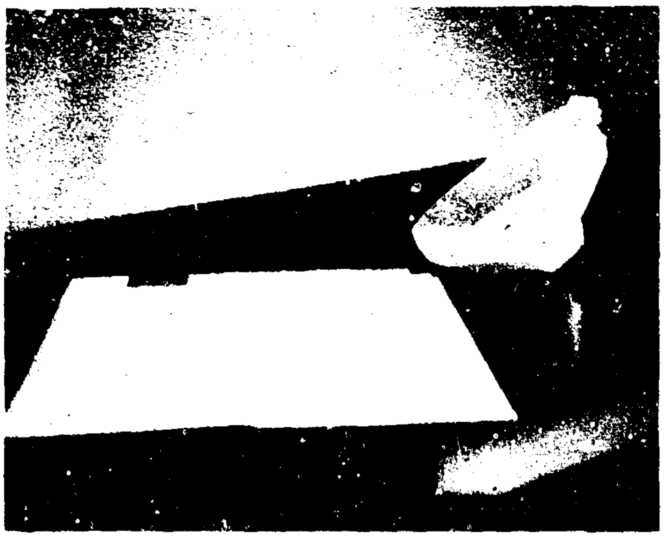
Micromerograph Particle Size Analysis

Particle-Size Computation

PRODUCTION SEQUENCE CONTINUED

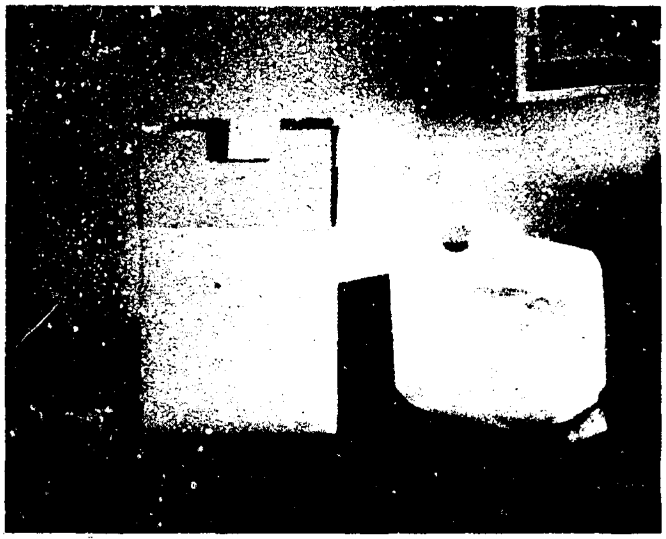
Pneumatic Bottle
Filling Apparatus

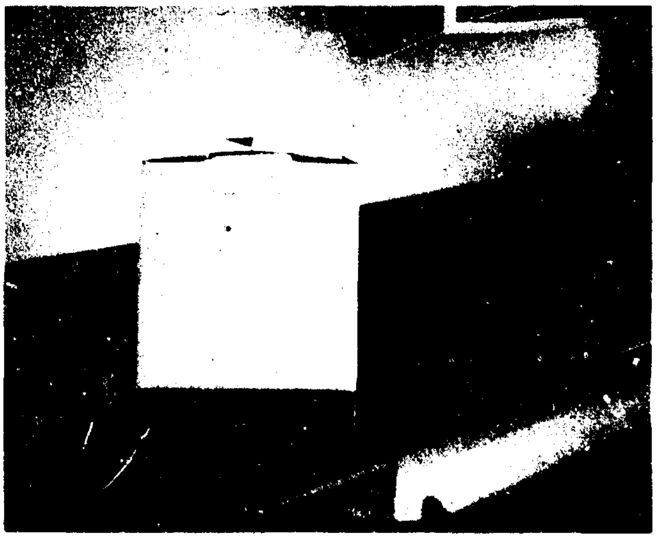
Pneumatic Loading of
Finished Product



Packaging Components

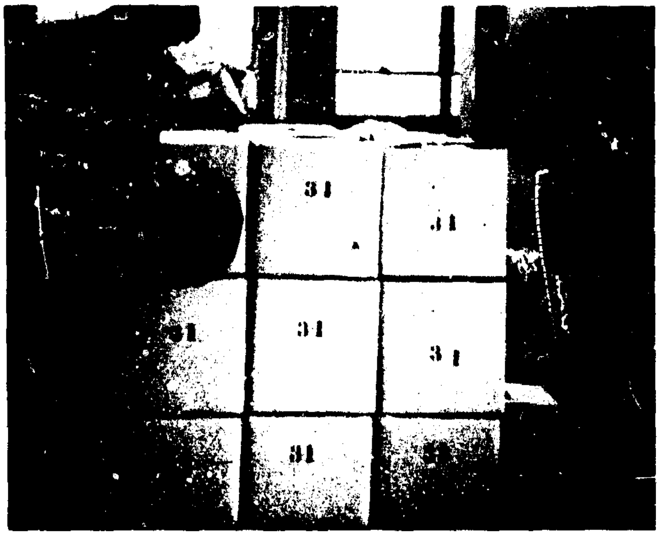
PRODUCTION SEQUENCE CONTINUED



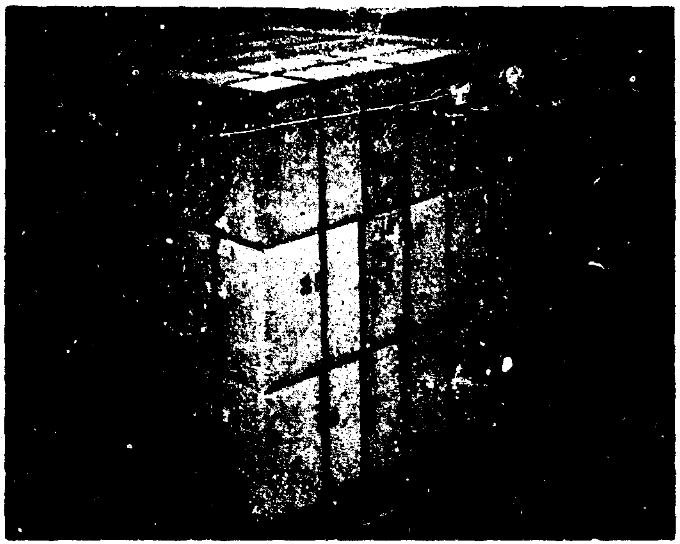


Finished Carton
Containing 25 lbs. of Product

PRODUCTION SEQUENCE CONTINUED

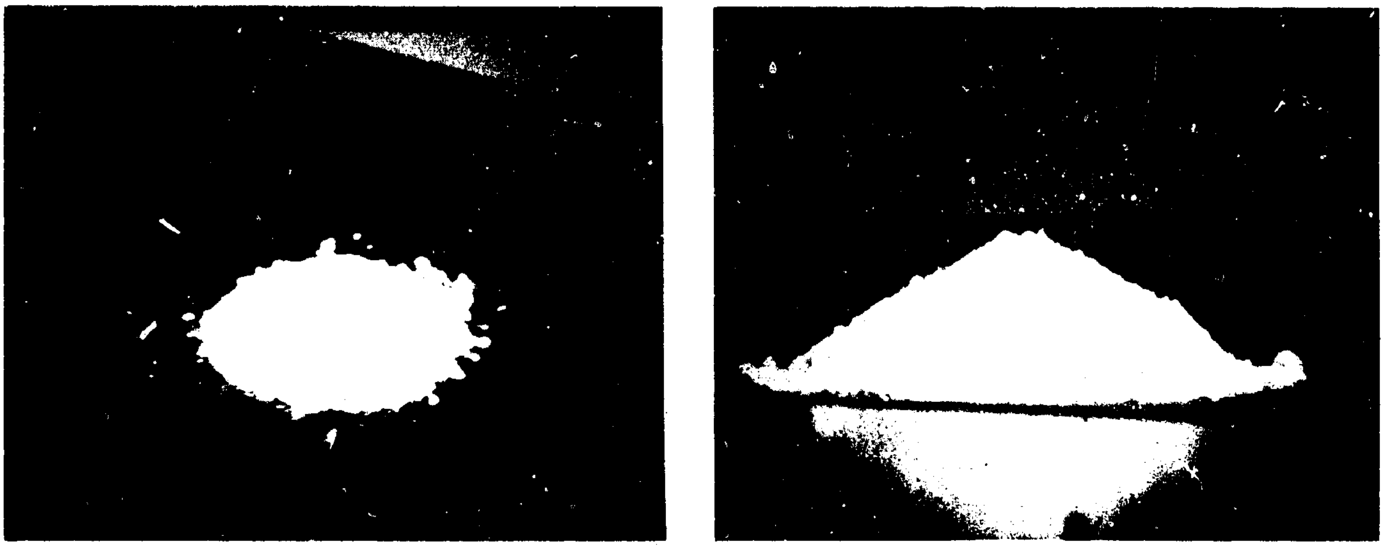


Labelling for Shipment

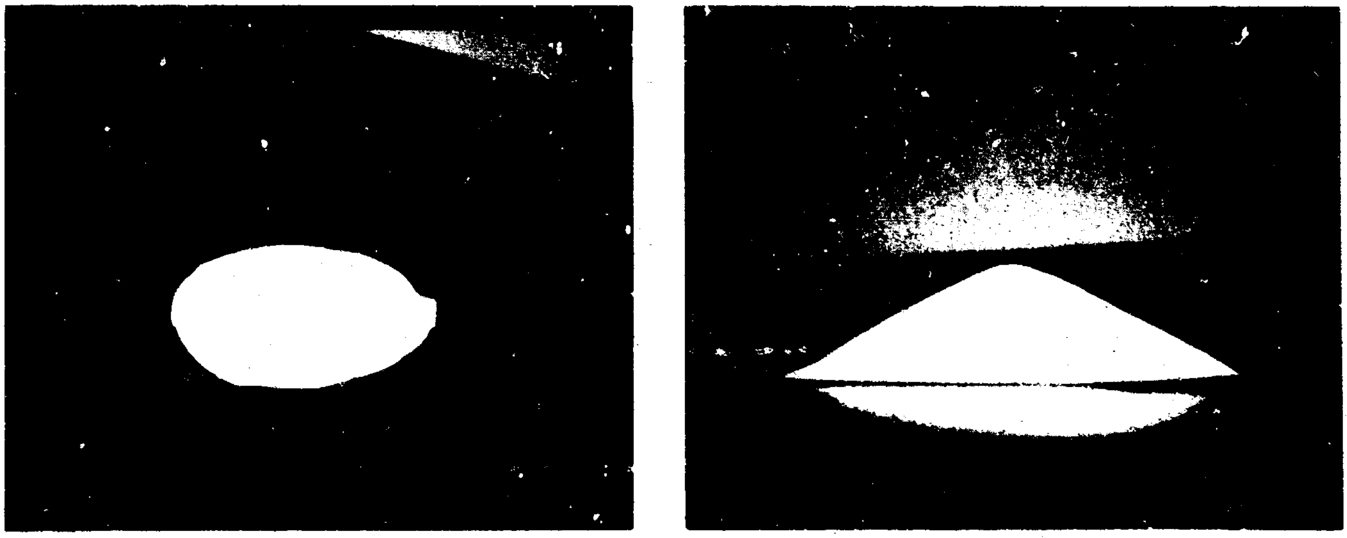


27-Carton Pallet for Delivery

UREA-BEFORE AND AFTER ENCAPSULATION



Finely-Ground Urea Raw Material



Urea After Ethylcellulose Encapsulation

APPENDIX II

PARTICLE NUMBER DISTRIBUTION OF ENCAPSULATED UREA PRODUCTION BATCHES

FREQUENCY IN NUMBER PERCENT

Size Range/Batch No.	AP-432	AP-433	AP-434	UC-4	UC-5	UC-6
159-238 μ	0.00%	0.00%	0.00%	0.03%	0.02%	0.35%
133-159	0.06	0.45	0.04	0.15	0.21	0.78
106-133	0.37	3.80	0.38	0.45	1.69	3.72
80-106	2.21	31.78	2.30	3.09	17.40	25.53
66-80	10.36	22.88	8.57	12.61	15.40	19.28
58-66	13.03	12.02	10.74	11.42	10.10	11.10
53-58	11.14	5.72	9.27	9.09	6.56	5.70
48-53	12.63	5.22	11.14	9.28	6.04	4.96
42-48	14.33	4.45	13.04	10.65	6.34	3.46
37-42	13.87	3.23	12.09	7.81	6.56	2.77
32-37	10.61	4.96	11.60	20.31	6.56	4.24
27-32	4.26	1.17	10.03	5.34	5.40	5.82
24-27	1.91	1.80	2.56	2.51	3.08	3.62
21-24	2.29	2.52	2.10	2.89	2.65	5.05
19-21	2.92	0.00	1.36	4.55	3.27	3.64
18-19	0.00		2.69	0.00	2.94	0.00
17-18			2.09		2.62	
16-17			0.00		2.00	
15-16					1.11	
1-15					0.00	
Mean Diameter, d_{av} =	65.77 μ	90.00 μ	65.32 μ	70.91 μ	81.62 μ	97.92 μ
Mean Volume Surface Diameter, d_{vs} =	59.89 μ	84.86 μ	58.74μ	62.22 μ	72.70 μ	87.98 μ

FREQUENCY IN NUMBER PERCENT

Size Range/Batch No.	UF-1	UF-2	UF-3	UF-4	UF-5	UF-6
159-238 μ	0.01%	0.00%	0.00%	0.00%	0.00%	0.00%
133-159	0.08	0.00	0.02	0.00	0.02	0.00
106-133	0.22	0.08	0.05	0.01	0.06	0.04
80-106	1.91	0.33	0.30	0.06	0.36	0.24
66-80	8.31	1.12	0.53	0.14	0.59	0.68
58-66	9.47	1.57	0.68	0.16	1.42	0.93
53-58	11.60	2.52	0.87	0.16	1.44	1.22
48-53	7.75	4.96	1.73	0.30	2.54	1.95
42-48	12.62	11.18	4.14	0.54	4.48	3.55
37-42	12.14	18.30	9.56	1.60	7.08	6.79
32-37	13.61	19.03	14.96	3.15	10.28	10.04
27-32	6.65	13.52	19.22	3.38	10.66	10.54
24-27	7.75	6.24	12.08	2.66	6.39	7.06
21-24	1.80	9.50	10.01	4.08	4.00	6.38
19-21	2.62	1.15	11.65	6.38	8.98	10.14
18-19	3.47	4.57	3.88	2.55	1.34	5.18
17-18	0.00	1.77	1.27	3.20	1.56	2.62
16-17		4.15	1.50	2.80	1.24	5.58
15-16		0.00	4.40	3.32	2.24	4.13
14-15			3.14	3.38	1.76	7.16
13-14			0.00	3.83	2.10	3.40
12-13				4.39	11.57	5.12
11-12				11.92	5.28	7.25
9-11				10.78	14.63	0.00
7-9				13.94	0.00	
5-7				17.24		
1-5				0.00		
Mean Diameter, d_{av} =	63.80 μ	48.14 μ	46.45 μ	41.96 μ	53.19 μ	48.58 μ
Mean Volume Surface Diameter, d_{vs} =	53.44 μ	40.52 μ	38.28 μ	29.32 μ	42.20 μ	39.12 μ

FREQUENCY IN NUMBER PERCENT

Size Range/Batch No.	UC-7	UC-8	UC-9	UC-10	UC-11
159-238 μ	0.00%	0.01%	0.00%	0.00%	0.00%
133-159	0.07	0.13	0.00	0.06	0.04
106-133	0.43	0.47	0.12	0.33	0.12
80-106	3.10	2.94	0.76	2.36	0.63
66-80	10.33	11.10	2.41	12.05	1.35
58-66	10.76	13.14	4.88	13.26	1.96
53-58	8.88	10.05	5.28	10.88	2.30
48-53	10.44	11.25	8.26	11.84	3.68
42-48	12.10	11.96	11.91	13.06	6.03
37-42	12.01	9.14	14.59	11.75	9.65
32-37	11.50	9.80	14.34	7.37	12.72
27-32	9.26	8.07	14.46	4.32	12.52
24-27	2.34	3.26	6.19	3.04	8.38
21-24	2.64	2.46	4.81	2.10	8.25
19-21	4.90	0.80	7.12	2.32	12.07
18-19	1.22	1.05	1.36	3.09	2.87
17-18	0.00	1.23	0.74	2.27	2.52
16-17		1.44	1.74	0.00	5.89
15-16		1.70	1.03		3.46
14-15		0.00	0.00		0.00
1-14					
Mean Diameter, d_{av} =	68.04 μ	69.64 μ	54.96 μ	66.35 μ	56.32 μ
Mean Volume Surface Diameter, d_{vs} =	60.96 μ	61.74 μ	48.61 μ	60.26 μ	45.96 μ

FREQUENCY IN NUMBER PERCENT

Size Range/Batch No.	UC-12	UC-13	UC-14	UC-15	UC-17
159-238 μ	0.00%	0.01%	0.00%	0.02%	0.16%
133-159	0.03	0.05	0.05	0.14	0.39
106-133	0.09	0.17	0.16	0.79	1.68
80-106	0.34	1.04	0.85	4.58	6.61
66-80	0.66	2.53	1.82	6.26	10.96
58-66	0.87	3.13	2.32	5.98	8.68
53-58	0.79	3.12	2.19	5.00	7.53
48-53	1.16	4.73	3.21	6.56	9.25
42-48	1.92	7.27	5.68	7.72	9.64
37-42	2.74	9.79	7.20	9.22	10.86
32-37	4.69	13.93	9.96	10.44	6.92
27-32	6.23	13.93	9.91	9.34	11.41
24-27	3.59	7.31	7.48	3.58	5.21
21-24	3.54	6.77	8.44	5.97	4.36
19-21	4.66	9.82	9.30	5.05	6.36
18-19	2.97	2.76	3.37	5.20	0.00
17-18	2.00	4.48	5.25	5.60	
16-17	1.81	3.67	4.43	8.56	
15-16	2.61	3.67	5.29	0.00	
14-15	2.44	1.83	4.22		
13-14	3.00	0.00	2.56		
12-13	7.95		6.32		
11-12	7.95		0.00		
9-11	10.51				
7-9	21.12				
5-7	6.35				
1-5	0.00				
Mean Diameter, d_{av} =	61.25 μ	60.97 μ	58.90 μ	77.47 μ	86.38 μ
Mean Volume Surface Diameter, d_{vs} =	43.29 μ	49.85 μ	46.18 μ	64.80 μ	72.11 μ

FREQUENCY IN NUMBER PERCENT

Size Range/Batch No.	UC-18	UC-19	UC-20	UC-21	UC-22	UC-23	UC-24
159-238 μ	0.07%	0.04%	0.00%	0.24%	0.16%	0.06%	0.02%
133-159	0.18	0.08	0.02	0.69	0.36	0.20	0.09
106-133	0.72	0.35	0.08	3.79	2.39	1.34	0.26
80-106	3.78	2.32	0.49	17.94	13.96	7.07	1.88
66-80	7.93	4.45	1.37	15.82	16.88	10.12	5.19
58-66	8.34	4.21	1.37	10.24	14.67	7.94	6.04
53-58	7.78	4.23	1.65	5.54	7.64	6.18	5.28
48-53	12.79	5.10	2.22	5.11	7.93	7.28	7.24
42-48	6.61	7.14	3.70	4.72	7.75	8.47	9.79
37-42	11.64	9.14	4.83	2.82	3.22	7.70	11.49
32-37	11.38	11.82	5.51	4.46	3.72	6.40	11.30
27-32	10.26	12.88	10.04	5.42	4.09	5.45	12.82
24-27	4.84	6.15	4.38	5.72	6.35	3.18	4.52
21-24	3.87	5.74	5.44	3.99	4.43	4.19	2.53
19-21	4.21	9.87	2.97	5.81	6.45	7.54	11.03
18-19	5.61	5.46	2.62	7.68	0.00	6.16	4.86
17-18	0.00	5.10	0.76	0.00		4.96	5.65
16-17		5.82	0.89			5.80	0.00
15-16			1.05				
14-15			1.25				
13-14			1.50				
12-13			9.15				
11-12			2.57				
9-11			4.24				
7-9			15.47				
5-7			16.44				
			0.00				
Mean Diameter, d_{av} =	76.61 μ	72.06 μ	61.56 μ	96.56 μ	89.50 μ	83.60 μ	67.92 μ
Mean Volume Surface Diameter, d_{vs} =	64.28 μ	57.79 μ	47.46 μ	85.59 μ	79.20 μ	71.52 μ	56.74 μ

FREQUENCY IN NUMBER PERCENT

Size Range/Batch No	UC-25	UC-26	UC-27	UC-28	UC-29	UC-30	UC-31
159-238 μ	0.00%	0.01%	0.00%	0.00%	0.00%	0.00%	0.00%
133-159	0.02	0.06	0.00	0.16	0.06	0.03	0.15
106-133	0.12	0.20	0.21	1.23	0.18	0.14	0.60
80-106	0.77	0.49	1.04	9.83	1.10	0.87	4.94
66-80	2.46	1.23	2.85	14.42	3.00	2.54	13.13
58-66	2.71	1.67	4.08	10.82	3.66	2.83	13.70
53-58	2.39	1.59	4.34	7.99	3.01	2.82	10.26
48-53	3.55	2.71	5.04	1.66	4.07	3.48	11.52
42-48	4.95	3.78	8.18	15.45	6.74	5.68	11.76
37-42	6.35	7.06	11.08	6.75	10.20	7.18	9.30
32-37	10.57	10.52	13.48	5.18	11.71	12.54	7.25
27-32	12.00	11.84	14.84	9.98	13.43	15.18	5.17
24-27	5.50	6.54	9.05	4.43	9.15	7.71	4.46
21-24	4.14	6.84	6.89	3.09	7.81	5.98	2.37
19-21	3.44	10.79	16.72	9.00	6.20	8.71	5.39
18-19	6.83	2.20	2.21	0.00	2.73	4.60	0.00
17-18	5.29	1.28	0.00		3.17	1.34	
16-17	6.19	1.49			3.72	3.13	
15-16	3.66	3.53			2.20	5.55	
14-15	4.36	2.10			7.86	4.41	
13-14	2.61	5.03			0.00	5.27	
12-13	3.18	6.14				0.00	
11-12	8.92	12.91					
12-11	0.00						
Mean Diameter, d_{av} =	60.36 μ	63.68 μ	58.76 μ	79.59 μ	62.51 μ	59.57 μ	72.15 μ
Mean Volume Surface Diameter, d_{vs} =	48.79 μ	46.97 μ	49.85 μ	71.46 μ	51.04 μ	48.61 μ	65.13 μ

FREQUENCY IN NUMBER PERCENT

Size Range/Batch No.	UC-34	UC-36	UC-37	UC-38
159-238 μ	0.08%	0.05%	0.03%	0.00%
133-159	0.51	0.50	0.24	0.05
106-133	4.67	0.96	0.66	0.21
80-106	37.32	0.88	0.90	1.12
66-80	24.88	1.11	1.73	3.15
58-66	11.80	1.31	1.93	4.48
53-58	5.15	1.47	2.64	3.47
48-53	2.98	2.54	3.56	6.04
42-48	3.46	4.28	5.35	8.85
37-42	5.04	6.62	9.05	11.49
32-37	1.55	8.26	15.30	14.61
27-32	2.56	13.15	14.69	13.56
24-27	0.00	8.68	9.27	7.80
21-24		9.22	6.97	5.44
19-21		11.52	15.93	9.51
18-19		5.54	3.83	2.10
17-18		2.06	2.23	2.43
16-17		3.53	2.61	5.70
15-16		7.03	3.08	0.00
14-15		4.98	0.00	
13-14		5.95		
12-13		0.00		
11-12				
10-11				
Mean Diameter, d_{av} =	92.69 μ	94.46 μ	77.38 μ	60.49 μ
Mean Volume Surface Diameter, d_{vs} =	87.56 μ	63 37 μ	55.37 μ	51.22 μ

FREQUENCY IN NUMBER PERCENT

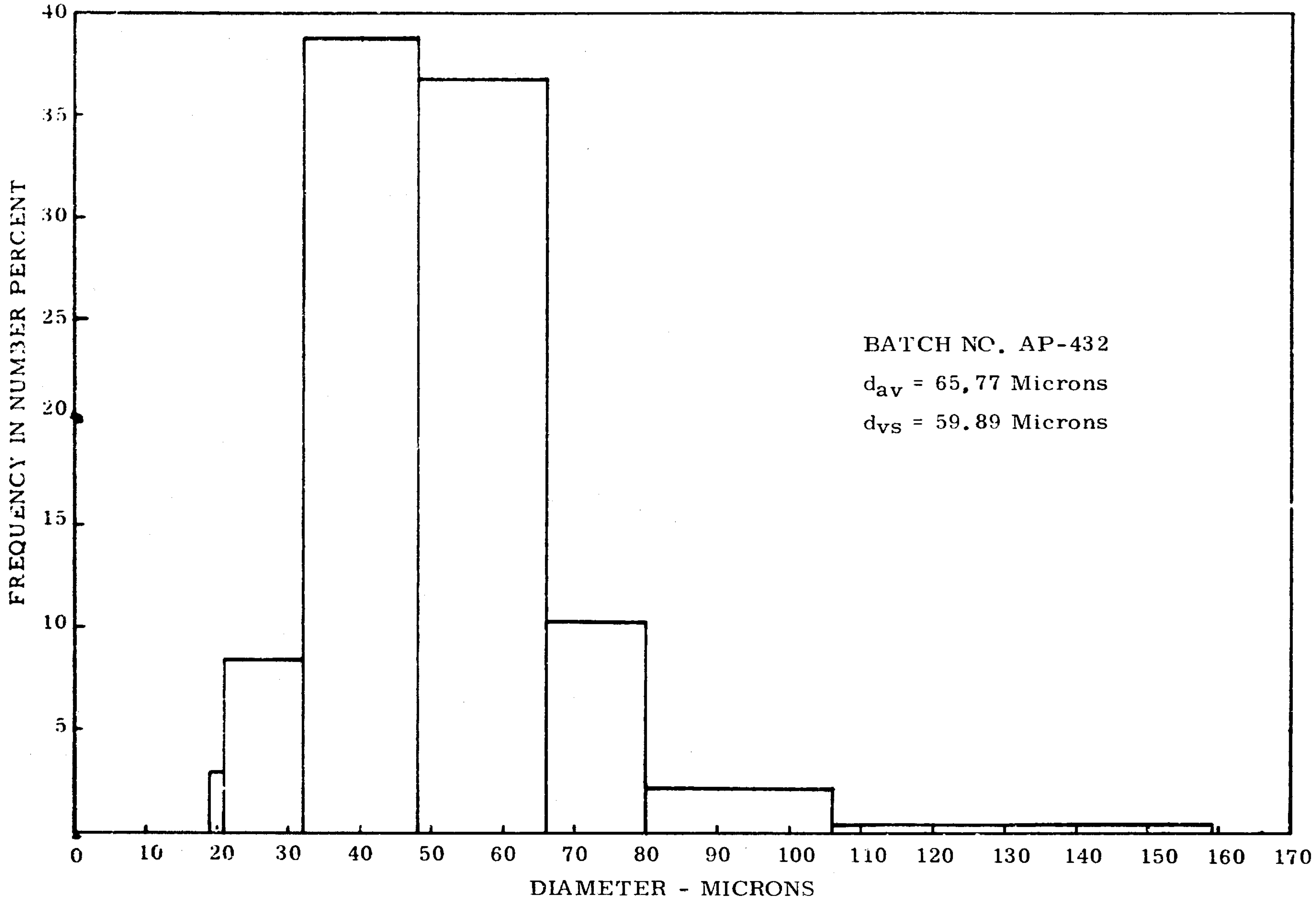
Size Range/Batch No.	UC-39	UC-40	UC-41	UC-42	UC-43	UC-44	UC-45
159-237 μ	0.00%	0.02%	0.10%	0.03%	0.00%	0.00%	0.12%
133-159	0.20	0.08	0.54	0.15	0.05	0.00	0.14
106-133	1.68	0.29	6.32	1.80	0.20	1.19	1.11
80-106	32.00	1.73	47.89	30.46	0.96	7.67	18.08
66-80	38.47	5.89	23.37	24.98	2.17	21.02	30.50
58-66	16.89	9.73	8.51	10.40	3.85	16.40	19.97
53-58	5.56	9.78	1.91	3.49	4.61	12.52	11.26
48-53	2.31	14.63	1.57	23.33	7.04	10.80	9.01
42-48	1.89	16.44	0.40	1.47	11.31	10.83	4.78
37-42	1.33	16.63	1.15	1.28	15.27	3.94	2.29
32-37	0.00	13.41	0.88	0.98	18.33	6.92	2.73
27-32		7.98	2.90	1.63	14.52	4.28	0.00
24-27		3.41	4.45	0.00	9.39	4.43	
21-24			0.00		7.87	0.00	
19-21					1.91		
18-19					2.52		
17-18					0.00		
Mean Diameter, d_{av} =	85.22 μ	64.55 μ	95.48 μ	85.27 μ	58.08 μ	74.89 μ	83.33 μ
Mean Volume Surface Diameter, d_{vs} =	82.32 μ	57.07 μ	91.58 μ	80.80 μ	49.55 μ	69.40 μ	77.41 μ

FREQUENCY IN NUMBER PERCENT

Size Range/Batch No.	UC-46	UC-47	UC-48	UC-49	UC-50	UC-51	UC-52
159-237 μ	0.15%	0.08%	0.11%	0.09%	0.00%	0.06%	0.00%
133-159	0.21	0.22	0.95	0.17	0.00	0.11	0.19
106-133	1.91	0.77	24.25	1.15	1.15	2.20	0.78
80-106	32.31	14.65	32.27	26.99	26.79	39.30	26.28
66-80	34.60	31.78	18.12	35.59	32.64	33.91	36.12
58-66	18.91	21.82	8.67	19.57	19.31	14.30	18.96
53-58	6.27	12.10	4.02	7.84	6.39	4.81	8.39
48-53	2.45	8.32	1.15	4.23	3.04	2.71	4.72
42-48	1.53	5.17	2.51	2.22	2.83	1.51	2.64
37-42	1.66	3.22	2.55	2.15	2.06	1.10	1.92
32-37	0.00	1.87	2.16	0.00	3.17	0.00	0.00
27-32		0.00	3.29		2.62		
24-27			0.00		0.00		
21-24							
19-21							
18-19							
17-18							
Mean Diameter, d_{av} =	88.09 μ	80.58 μ	82.37 μ	84.97 μ	82.73 μ	88.10 μ	82.60 μ
Mean Volume Surface Diameter, d_{vs} =	83.51 μ	75.36 μ	78.77 μ	80.68 μ	79.47 μ	84.73 μ	79.37 μ

FREQUENCY IN NUMBER PERCENT

Size Range/Batch No.	UC-53	UC-54	UC-55
159-237 μ	0.17%	0.00%	0.06%
133-159	0.20	0.00	0.15
106-133	1.23	0.11	0.59
80-106	23.32	1.34	7.31
66-80	37.33	4.45	27.94
58-66	17.49	8.82	22.91
53-58	11.01	8.54	14.01
48-53	4.32	12.14	10.64
42-48	2.01	15.68	8.25
37-42	2.92	16.34	3.20
32-37	0.00	14.68	4.92
27-32		13.29	0.00
24-27		1.21	
21-24		3.39	
19-21		0.00	
18-19			
17-18			
Mean Diameter, d_{av} =	86.04 μ	58.50 μ	76.10 μ
Mean Volume Surface Diameter, d_{vs} =	80.46 μ	53.39 μ	70.57 μ


BATCH NO. AP-432
d_{av} = 65,77 Microns
d_{vs} = 59.89 Microns
FREQUENCY IN NUMBER PERCENT
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140
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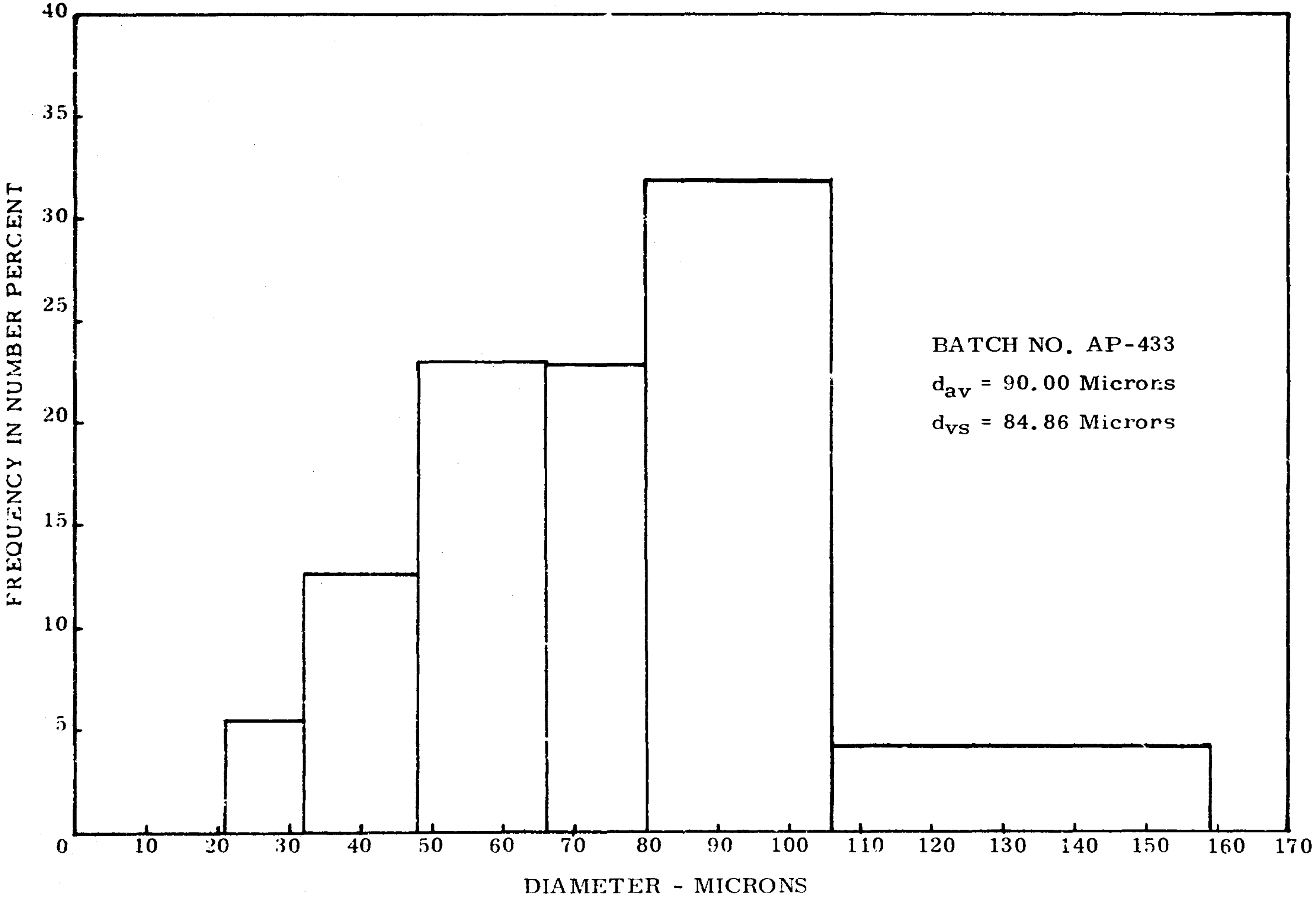
160
170
DIAMETER - MICRONS


BATCH NO. AP-433
d_{av} = 90.00 Microns
d_{vs} = 84.86 Microns
FREQUENCY IN NUMBER PERCENT
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5
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20
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30
35
40
DIAMETER - MICRONS
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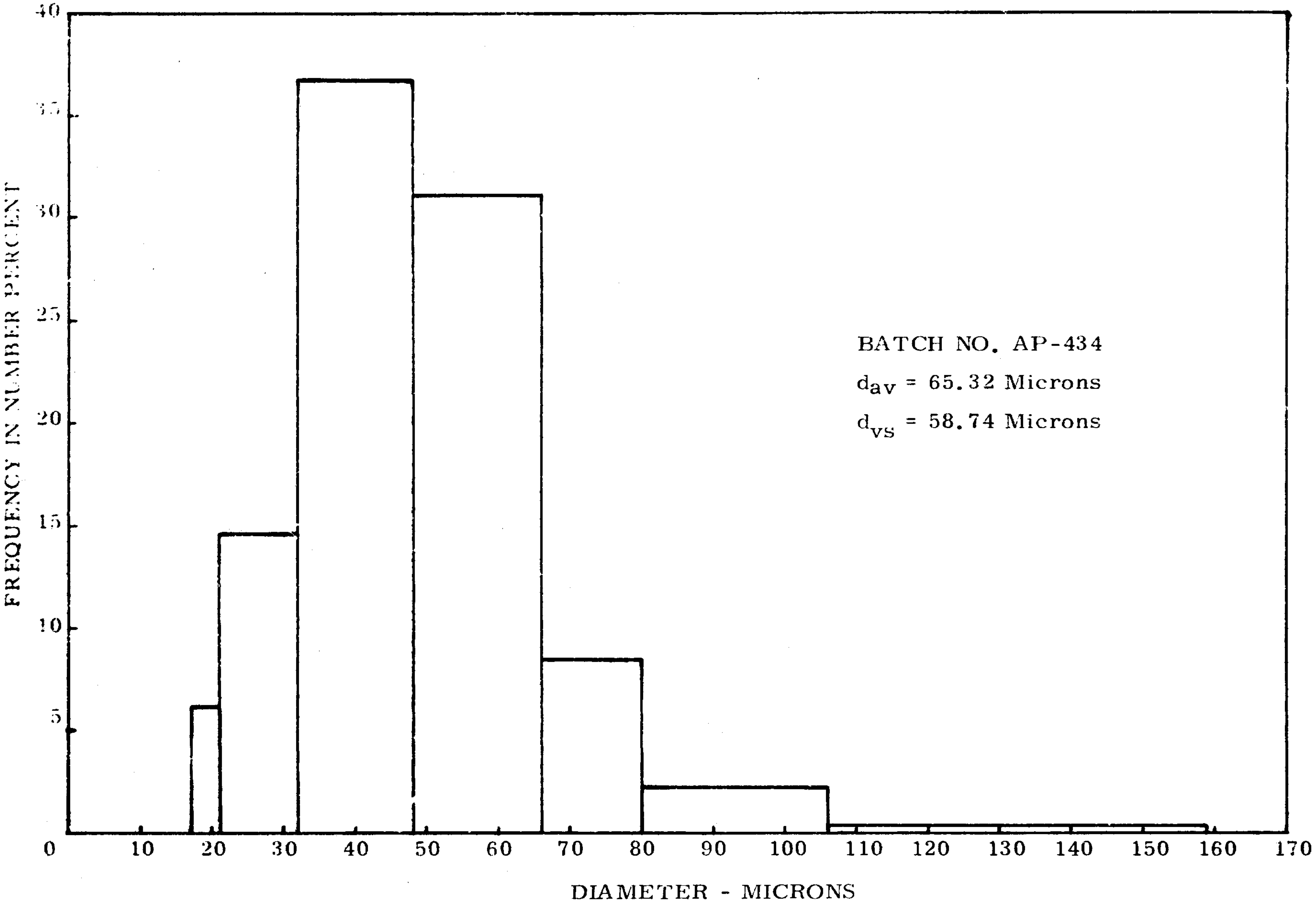
170


BATCH NO. AP-434
d_{av} = 65.32 Microns
d_{vs} = 58.74 Microns
FREQUENCY IN NUMBER PERCENT
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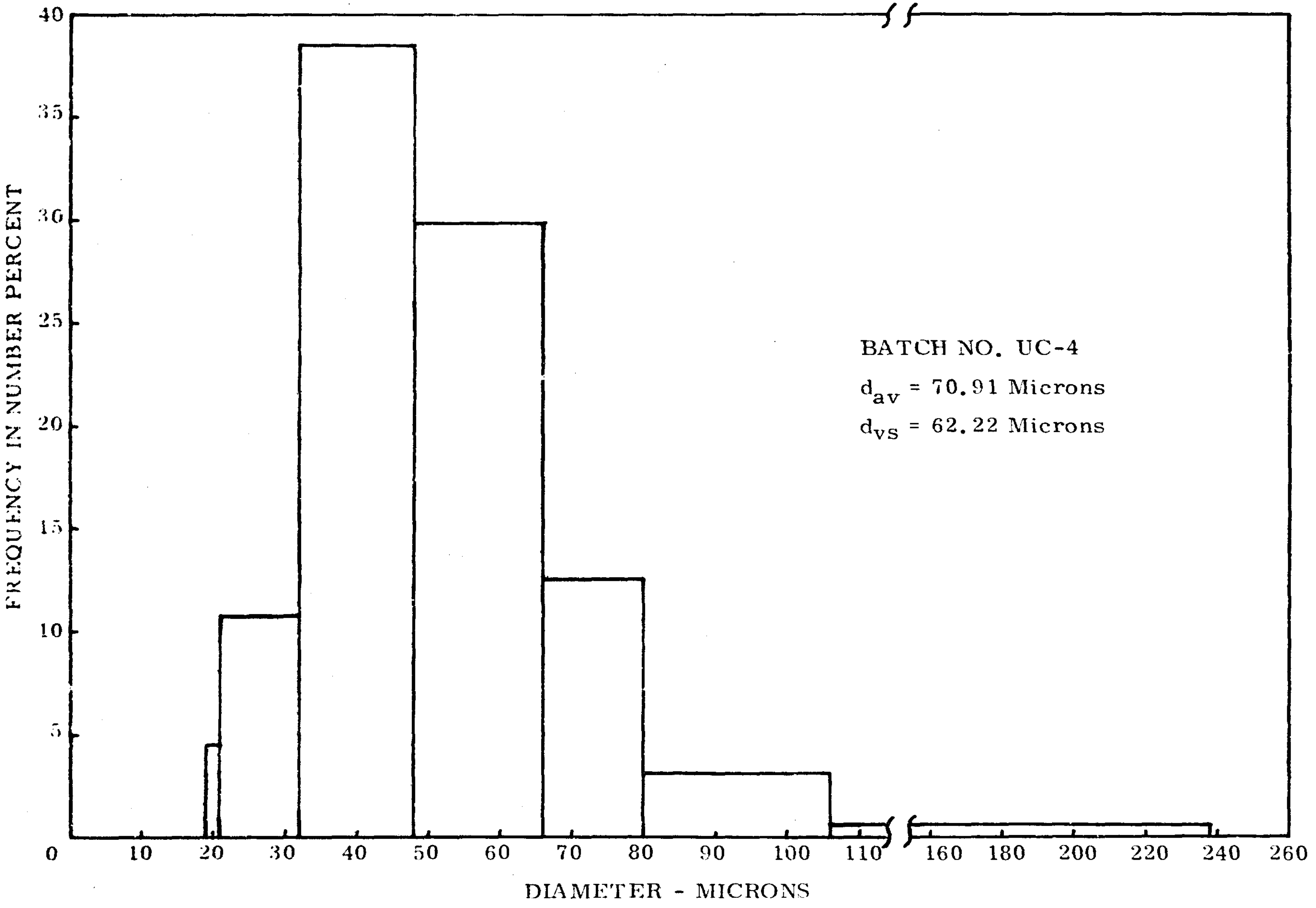
170
DIAMETER - MICRONS


BATCH NO. UC-4
d_{av} = 70.91 Microns
d_{vs} = 62.22 Microns
FREQUENCY IN NUMBER PERCENT
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220
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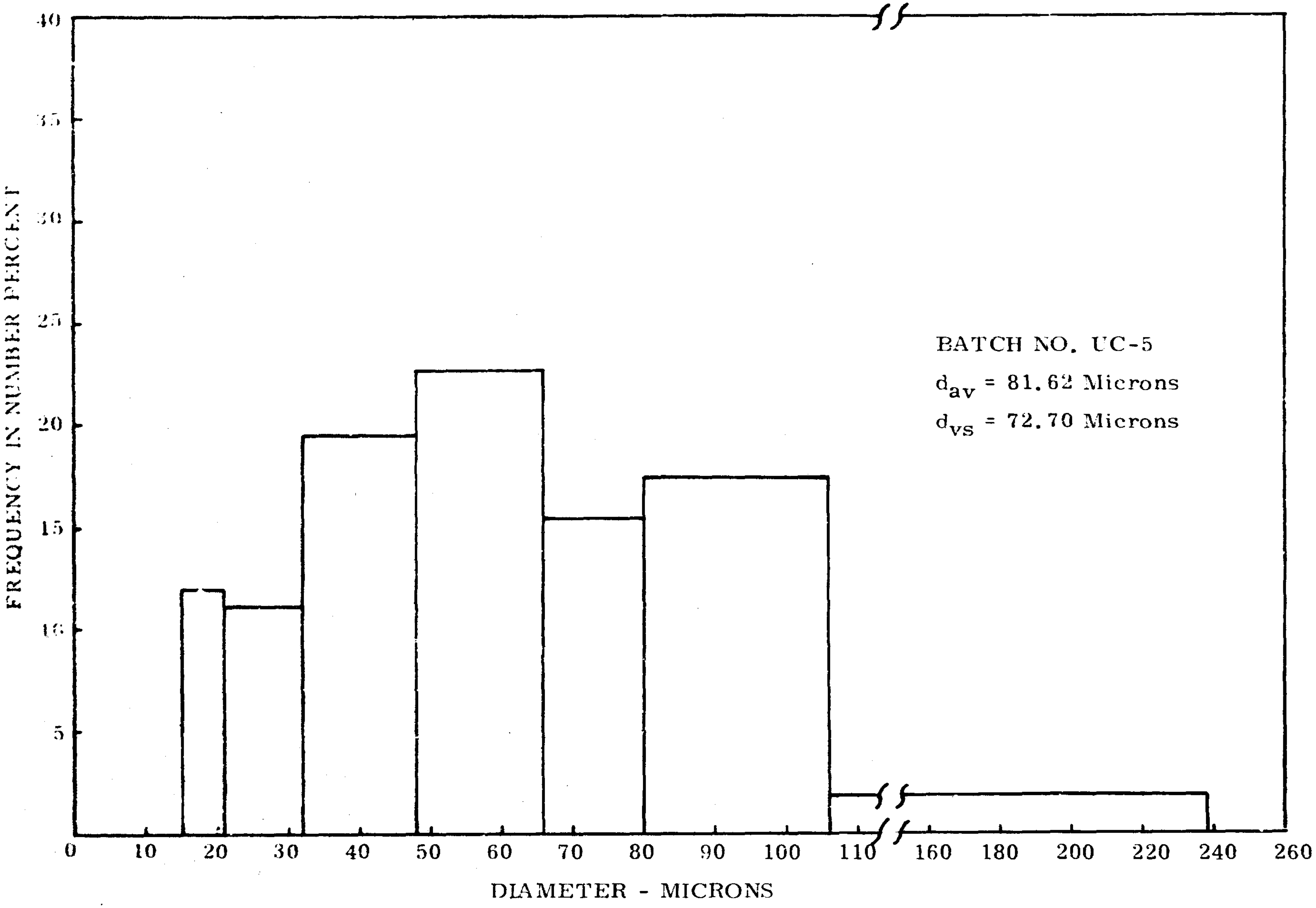
260
DIAMETER - MICRONS

BATCH NO. UC-5
d_{av} = 81.62 Microns
d_{vs} = 72.70 Microns
FREQUENCY IN NUMBER PERCENT
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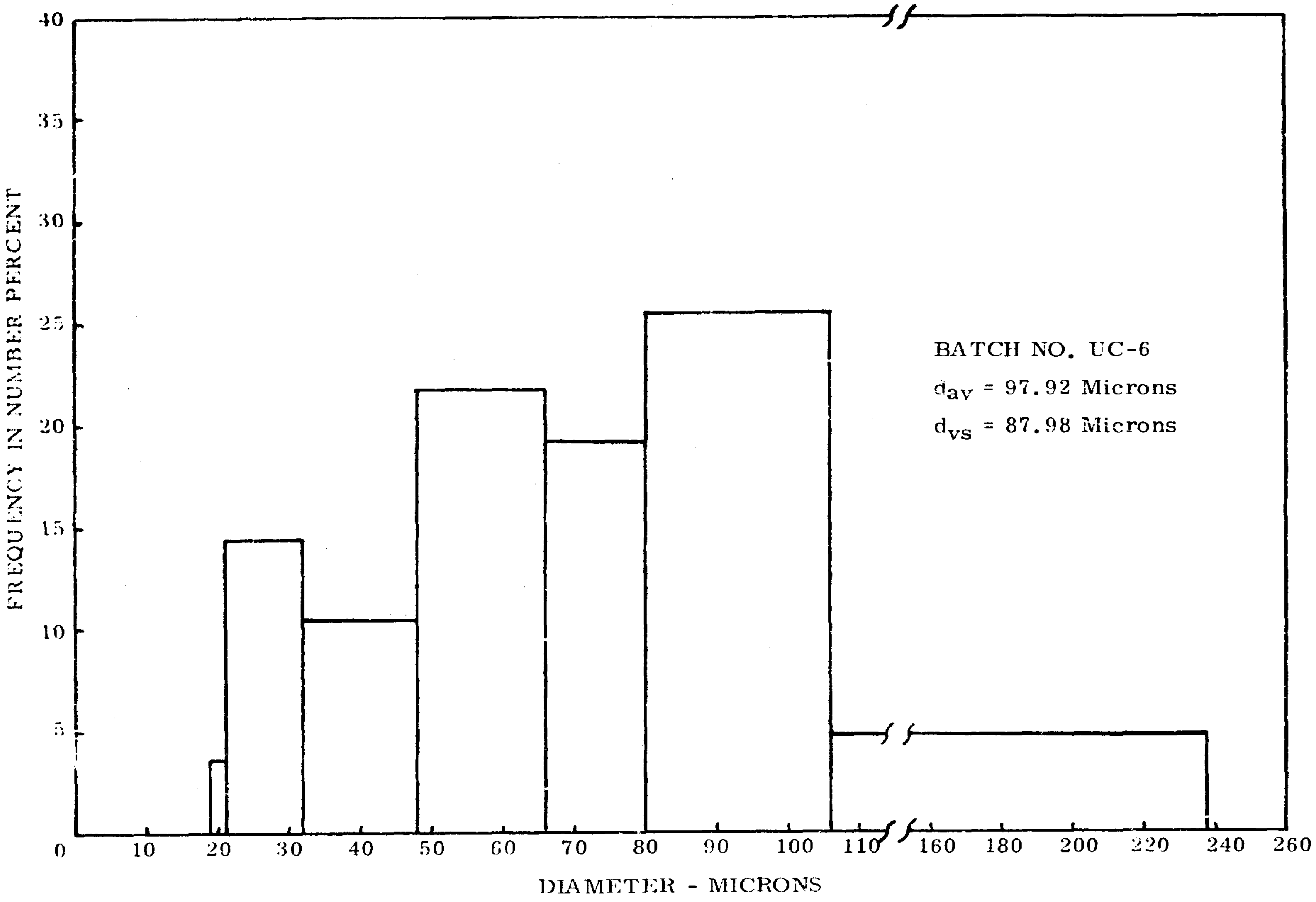
260
DIAMETER - MICRONS


BATCH NO. UC-6
d_{av} = 97.92 Microns
d_{vs} = 87.98 Microns
FREQUENCY IN NUMBER PERCENT
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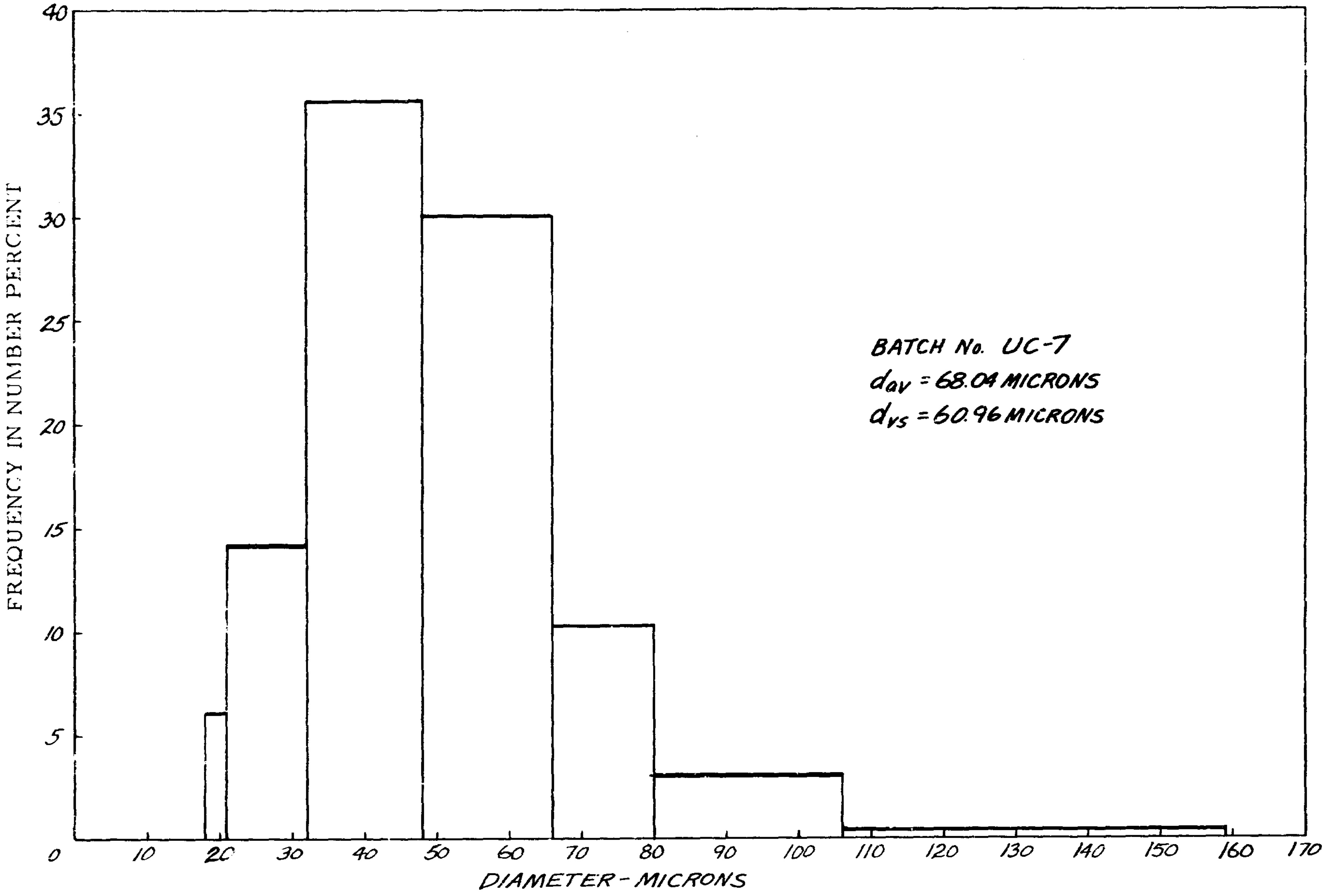
DIAMETER - MICRONS


BATCH No. UC-7
d_{av} = 68.04 MICRONS
d_{vs} = 60.96 MICRONS
FREQUENCY IN NUMBER PERCENT
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DIAMETER - MICRONS
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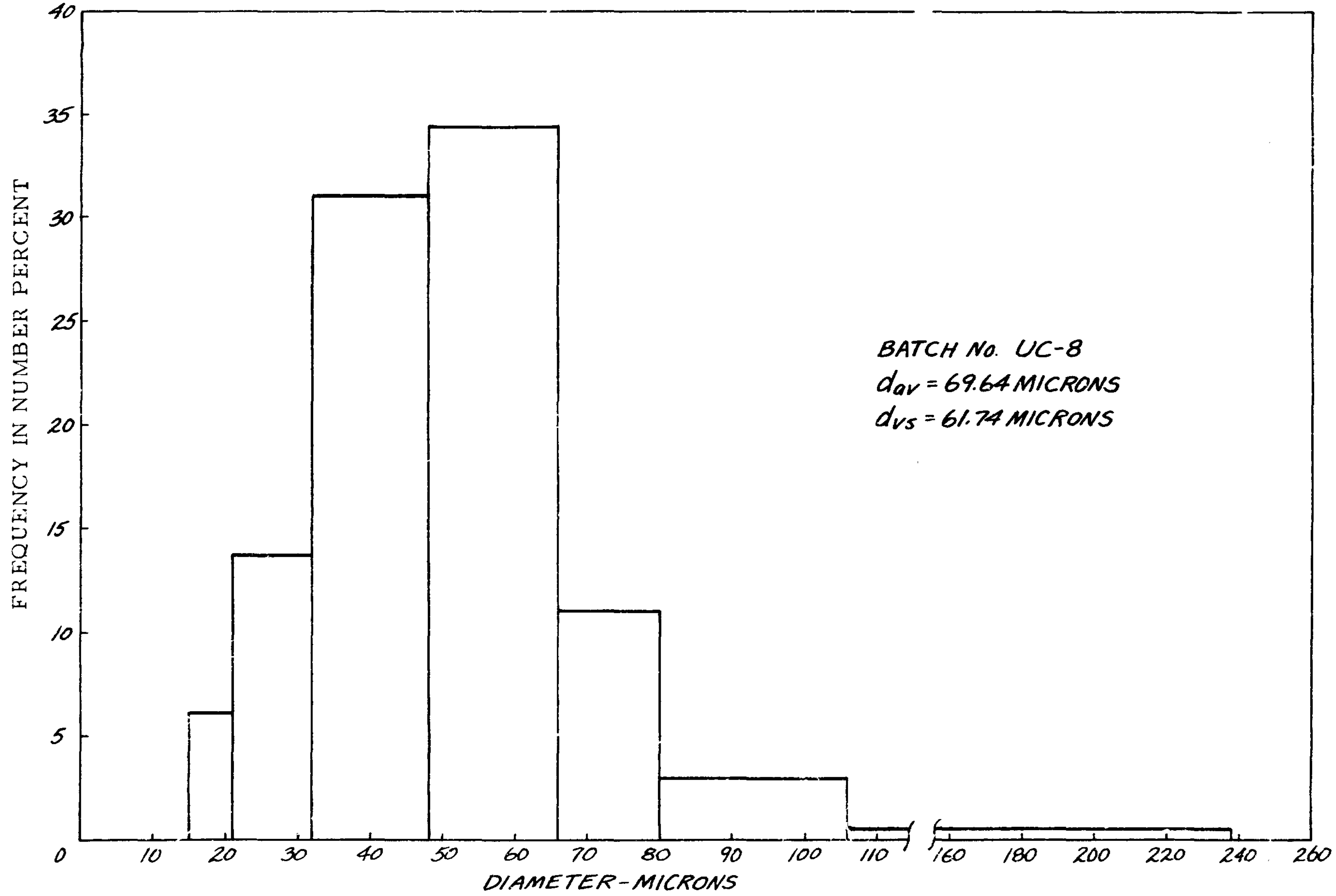
BATCH No. UC-8
d_{av} = 69.64 MICRONS
d_{vs} = 61.74 MICRONS
FREQUENCY IN NUMBER PERCENT
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40
DIAMETER-MICRONS
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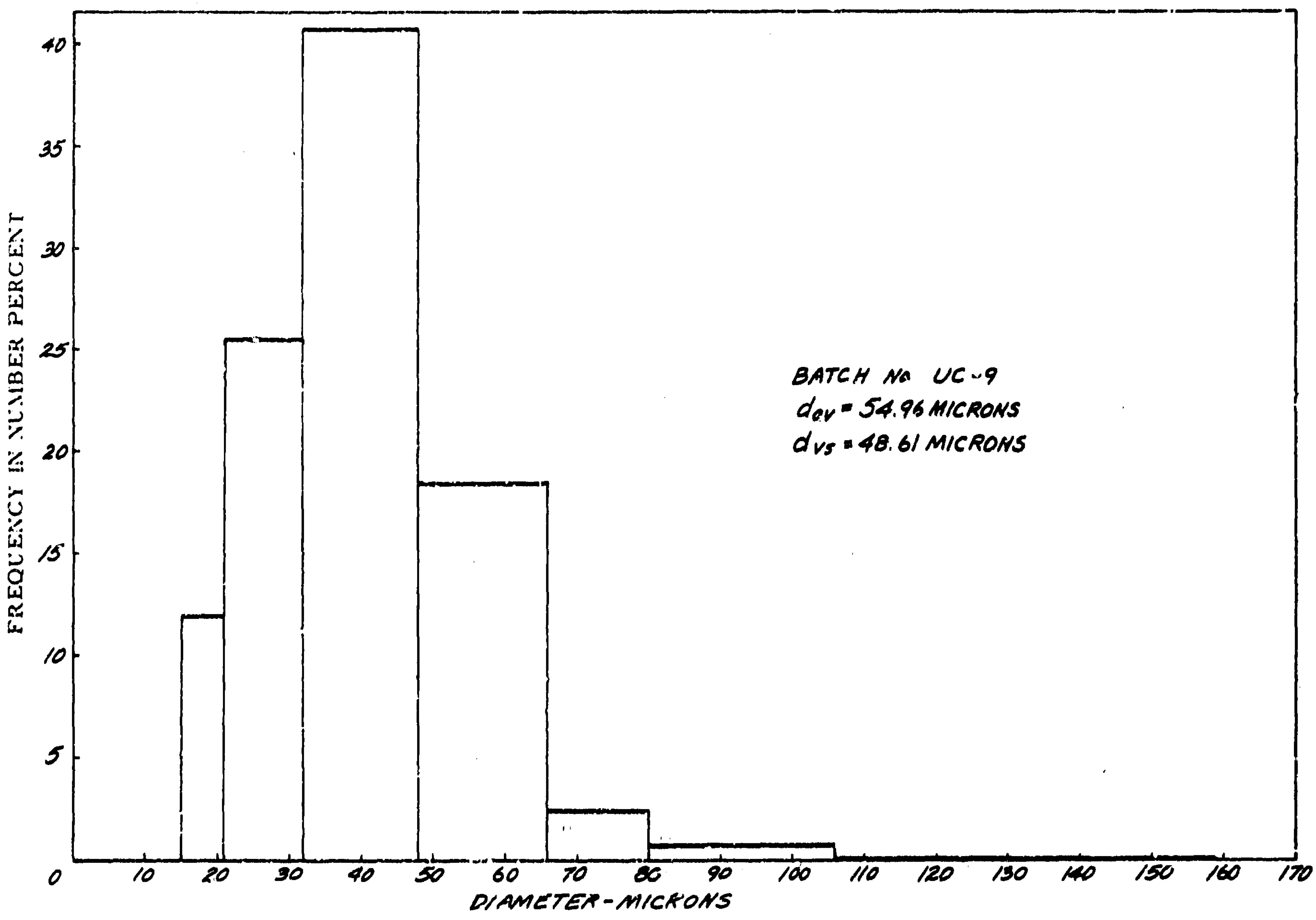
BATCH No UC-9
d_{ev} = 54.96 MICRONS
d_{vs} = 48.61 MICRONS
FREQUENCY IN NUMBER PERCENT
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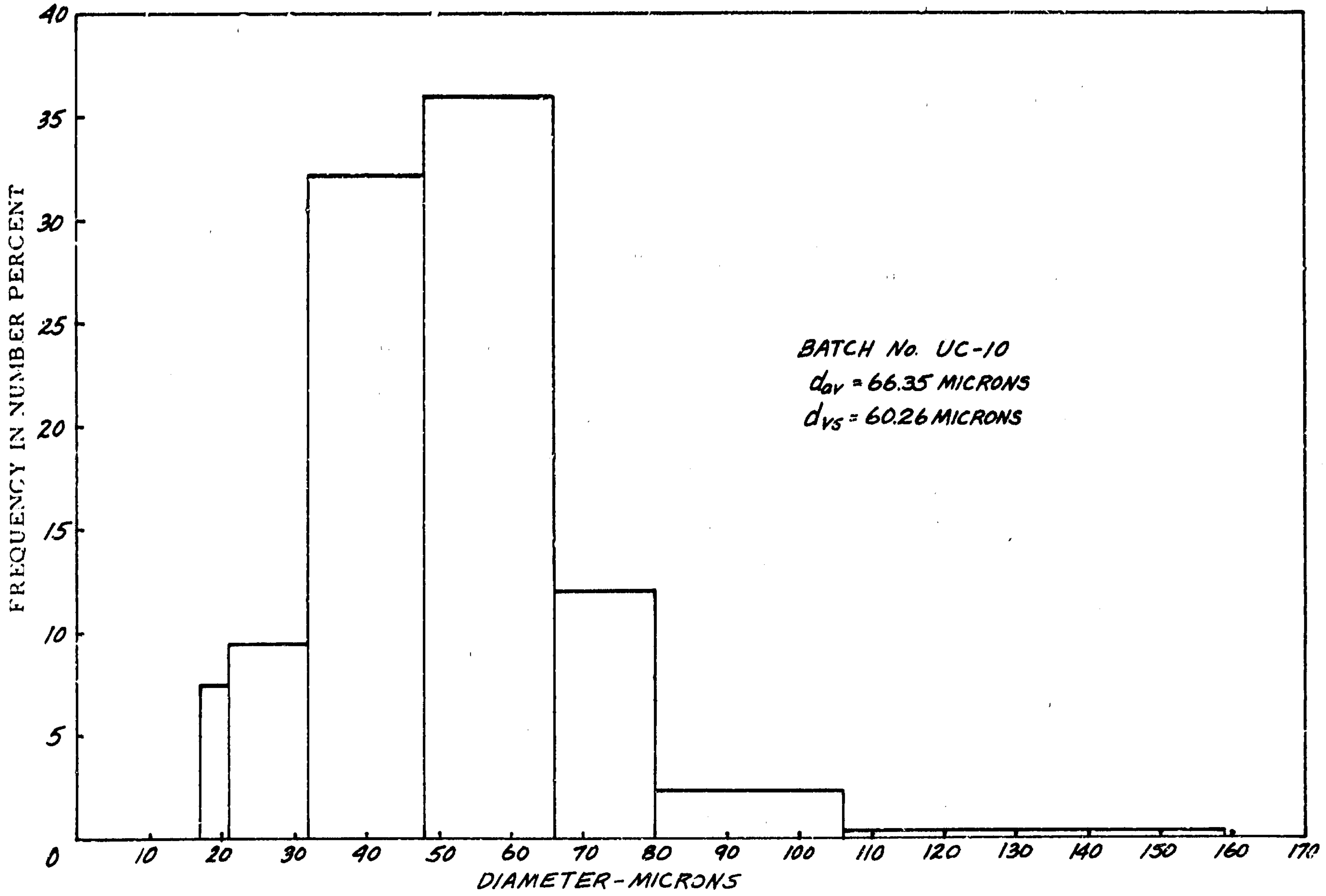
170
DIAMETER-MICRONS


BATCH No. UC-10
d_{av} = 66.35 MICRONS
d_{vs} = 60.26 MICRONS
FREQUENCY IN NUMBER PERCENT
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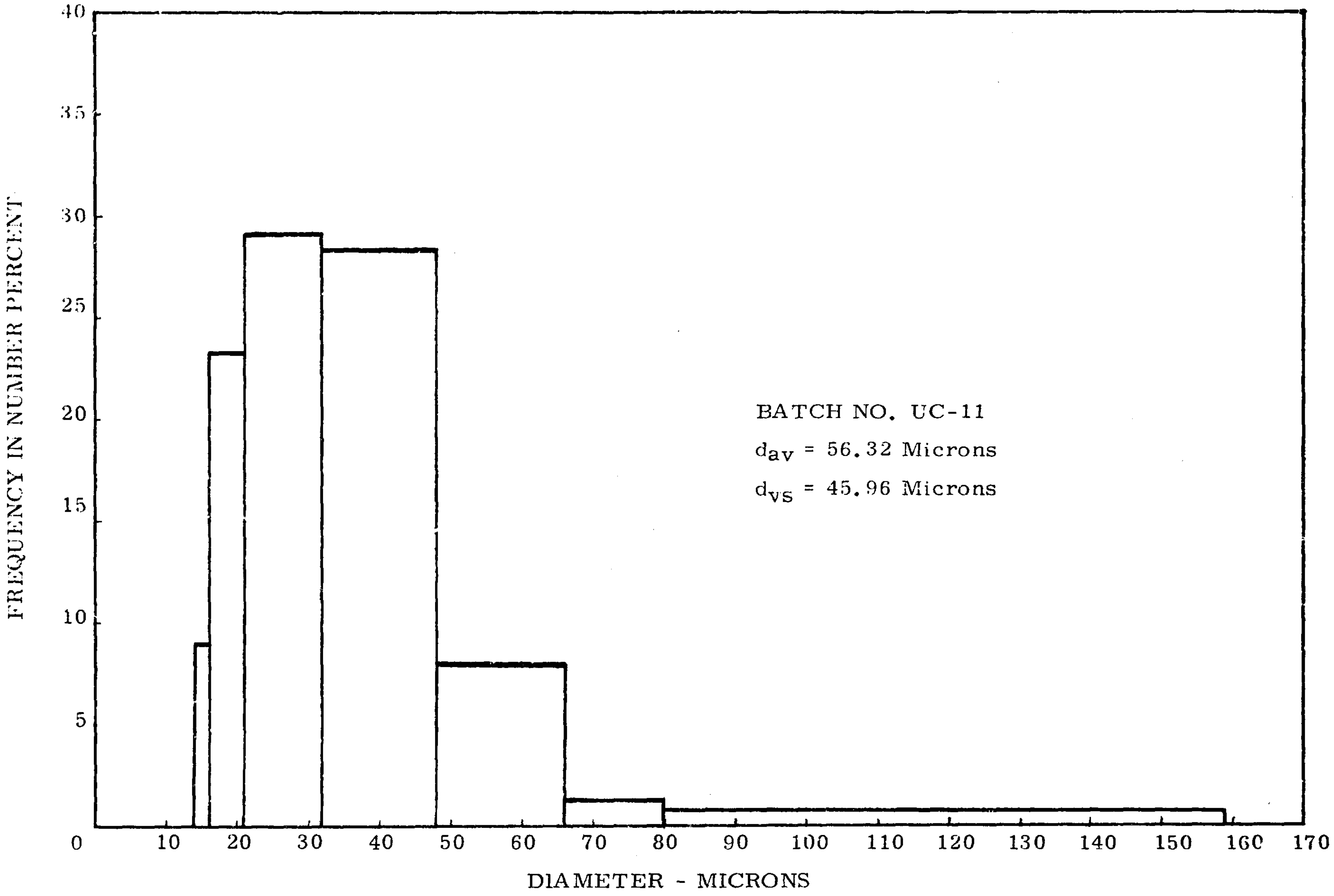
170
DIAMETER - MICRONS


BATCH NO. UC-11
d_{av} = 56.32 Microns
d_{vs} = 45.96 Microns
FREQUENCY IN NUMBER PERCENT
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15
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140
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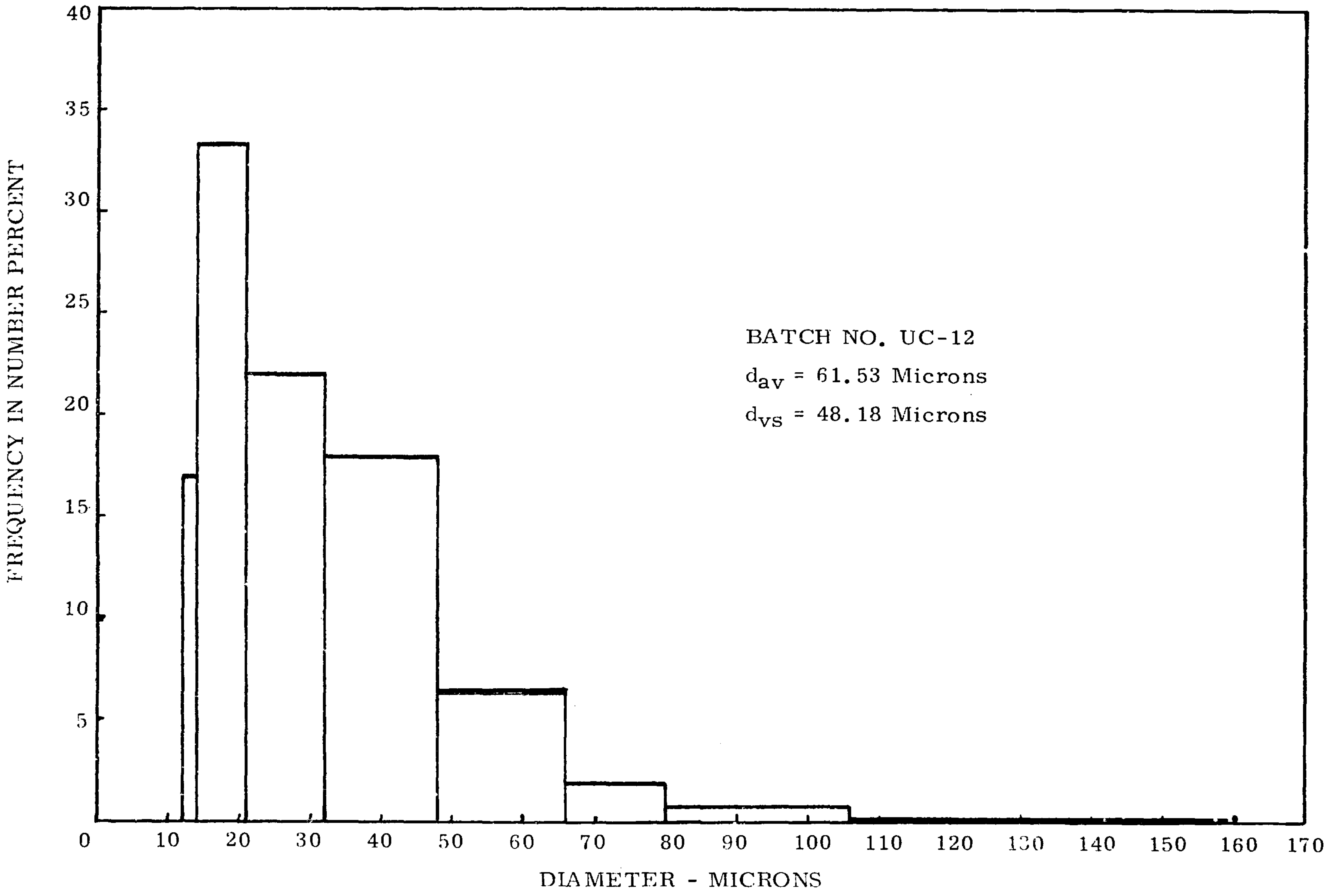
160
170
DIAMETER - MICRONS


BATCH NO. UC-12
d_av = 61.53 Microns
d_vs = 48.18 Microns
FREQUENCY IN NUMBER PERCENT
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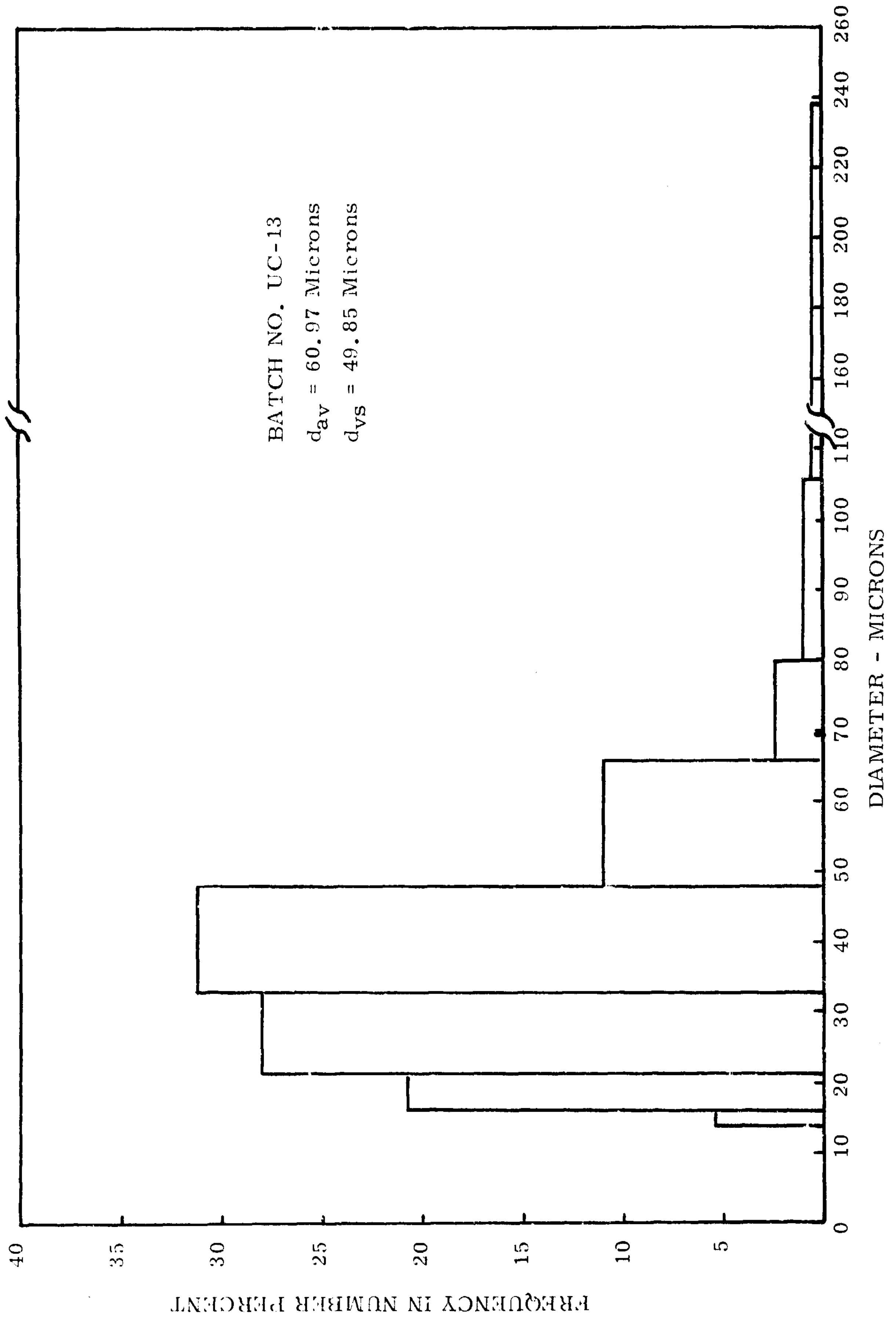
DIAMETER - MICRONS


BATCH NO. UC-13
d_{av} = 60.97 Microns
d_{vs} = 49.85 Microns
FREQUENCY IN NUMBER PERCENT
DIAMETER - MICRONS
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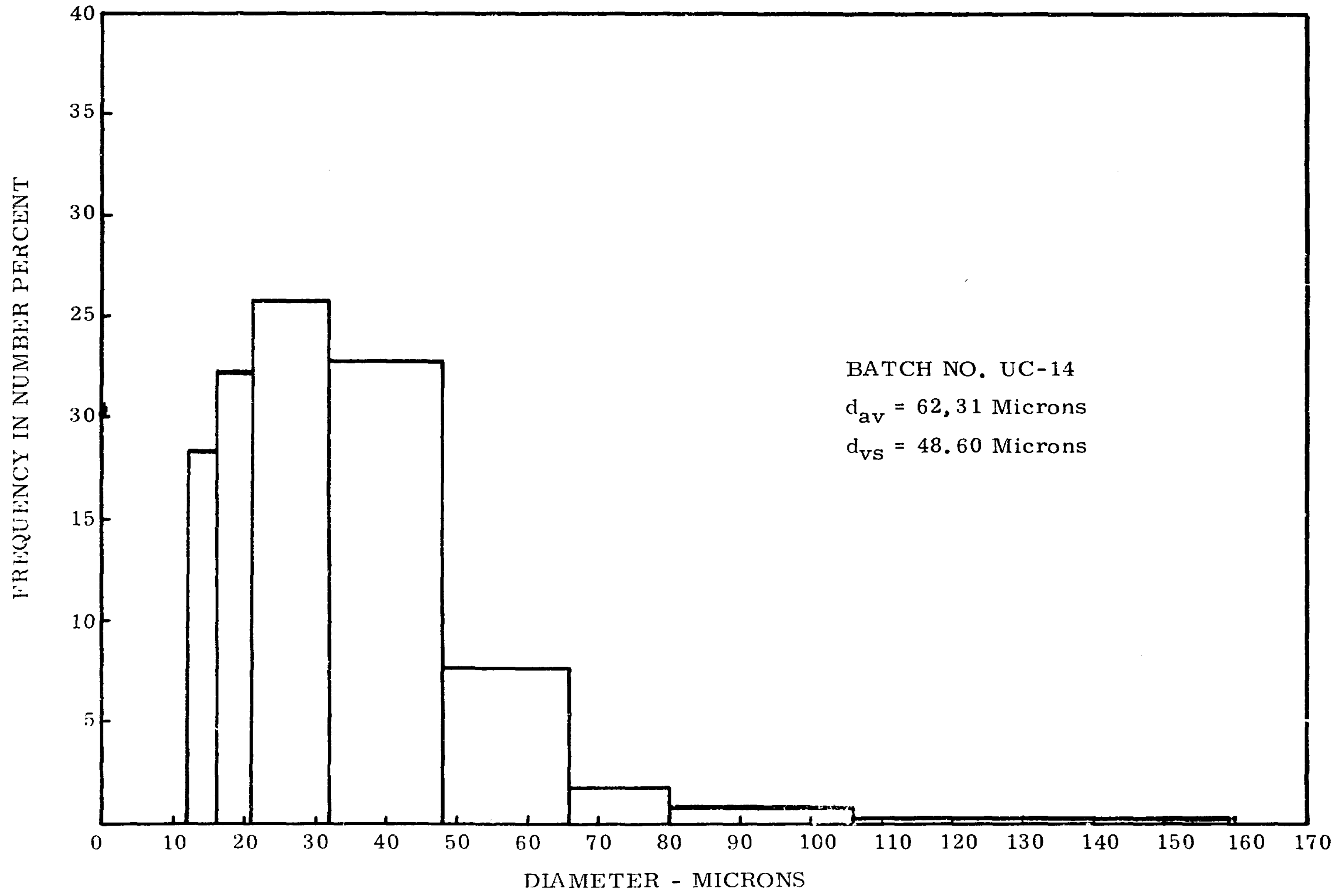
BATCH NO. UC-14
d_{av} = 62,31 Microns
d_{vs} = 48.60 Microns
FREQUENCY IN NUMBER PERCENT
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30
15
10
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0
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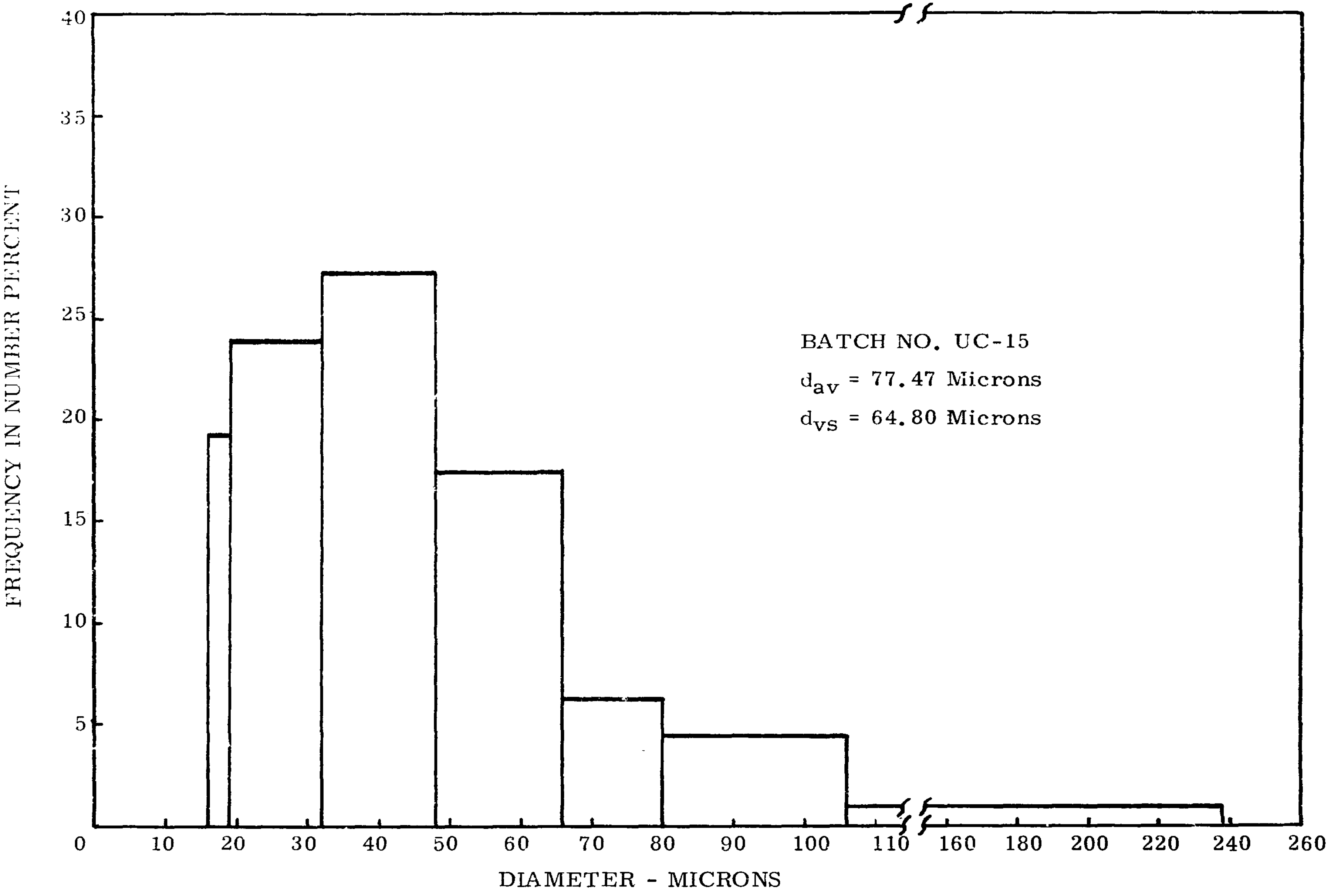
170
DIAMETER - MICRONS


BATCH NO. UC-15
d_{av} = 77.47 Microns
d_{vs} = 64.80 Microns
FREQUENCY IN NUMBER PERCENT
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15
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5
0
10
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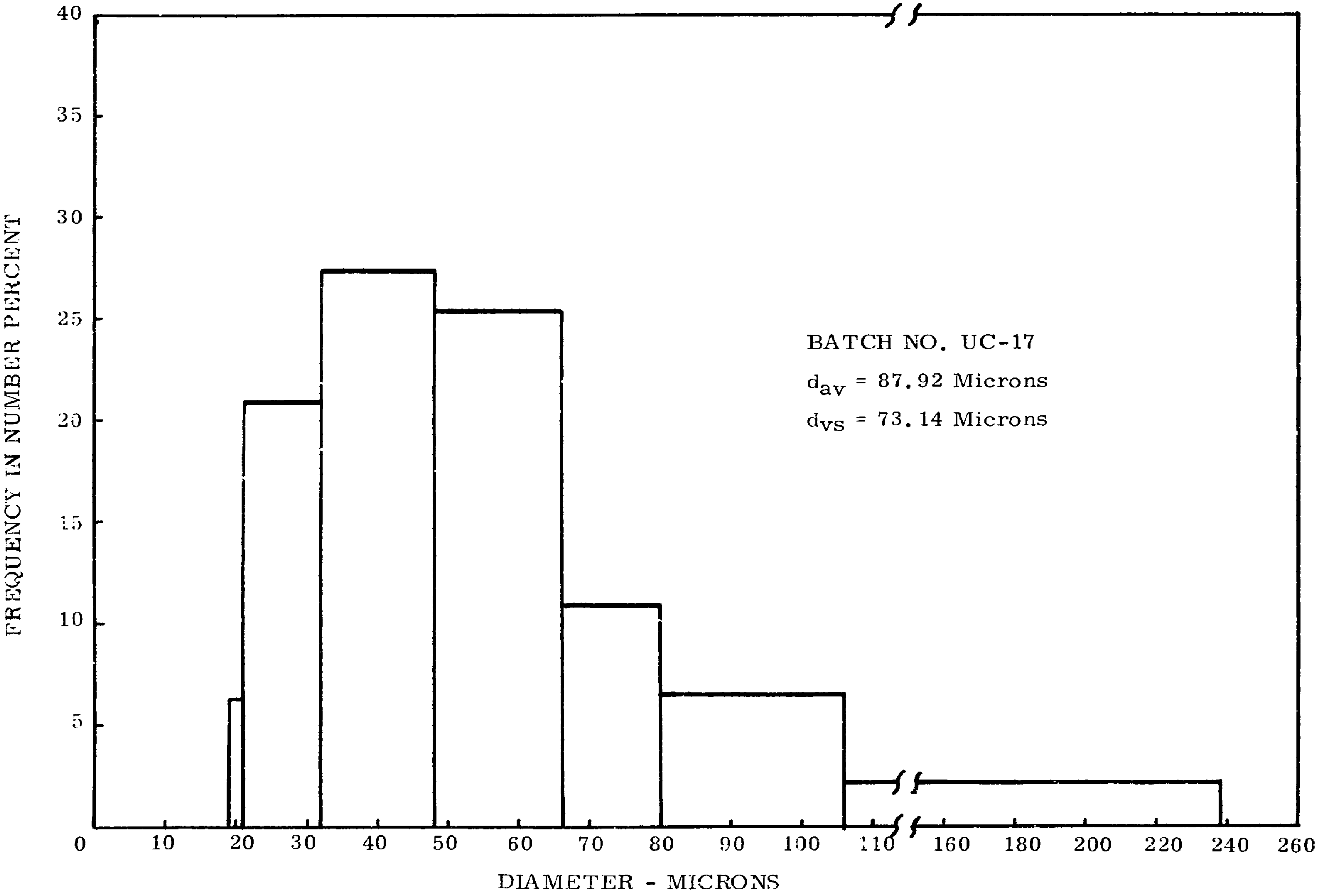
260
DIAMETER - MICRONS


BATCH NO. UC-17
d_{av} = 87.92 Microns
d_{vs} = 73.14 Microns
FREQUENCY IN NUMBER PERCENT
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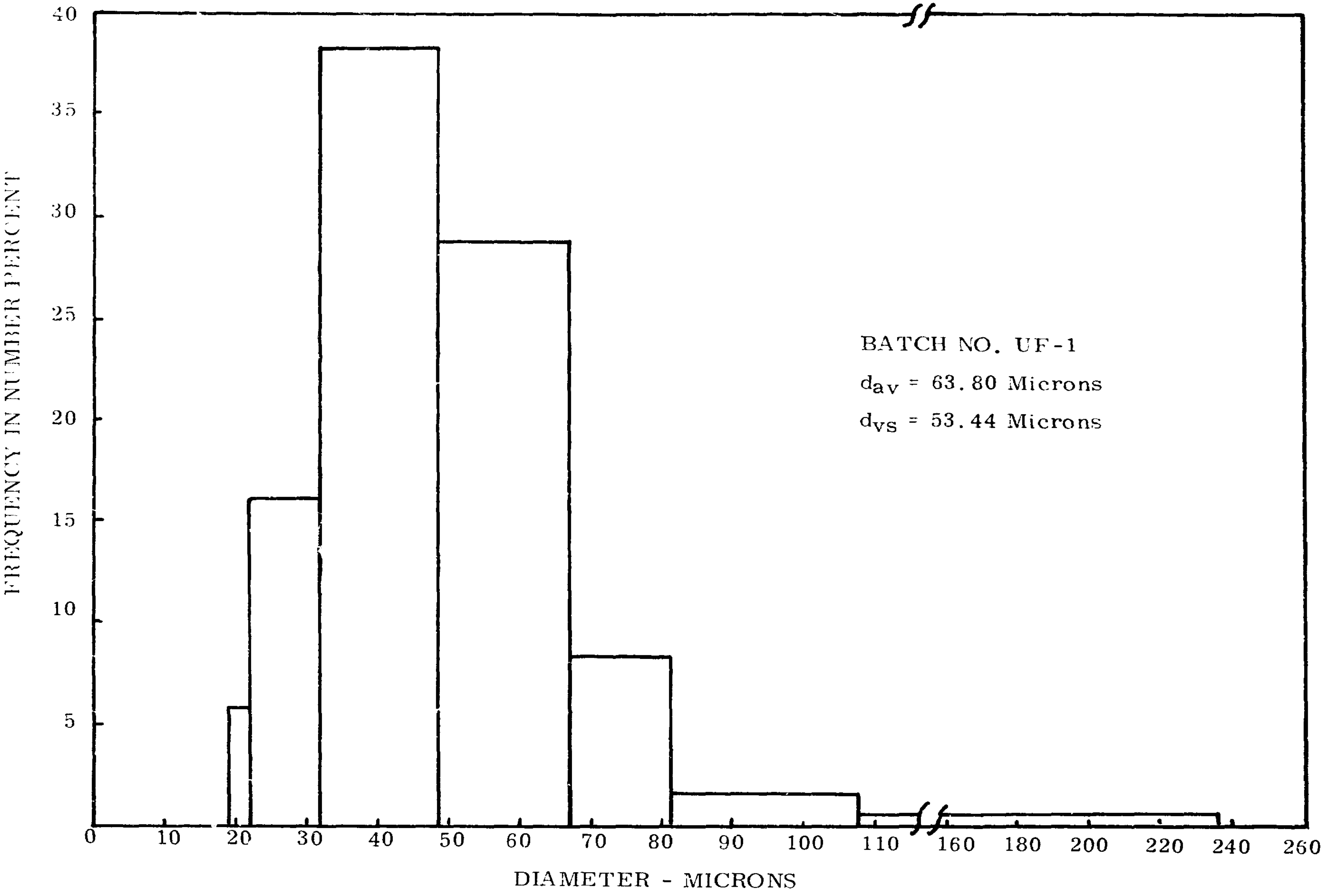
260
DIAMETER - MICRONS


BATCH NO. UF-1
d_{av} = 63.80 Microns
d_{vs} = 53.44 Microns
FREQUENCY IN NUMBER PERCENT
DIAMETER - MICRONS
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5
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15
20
25
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0
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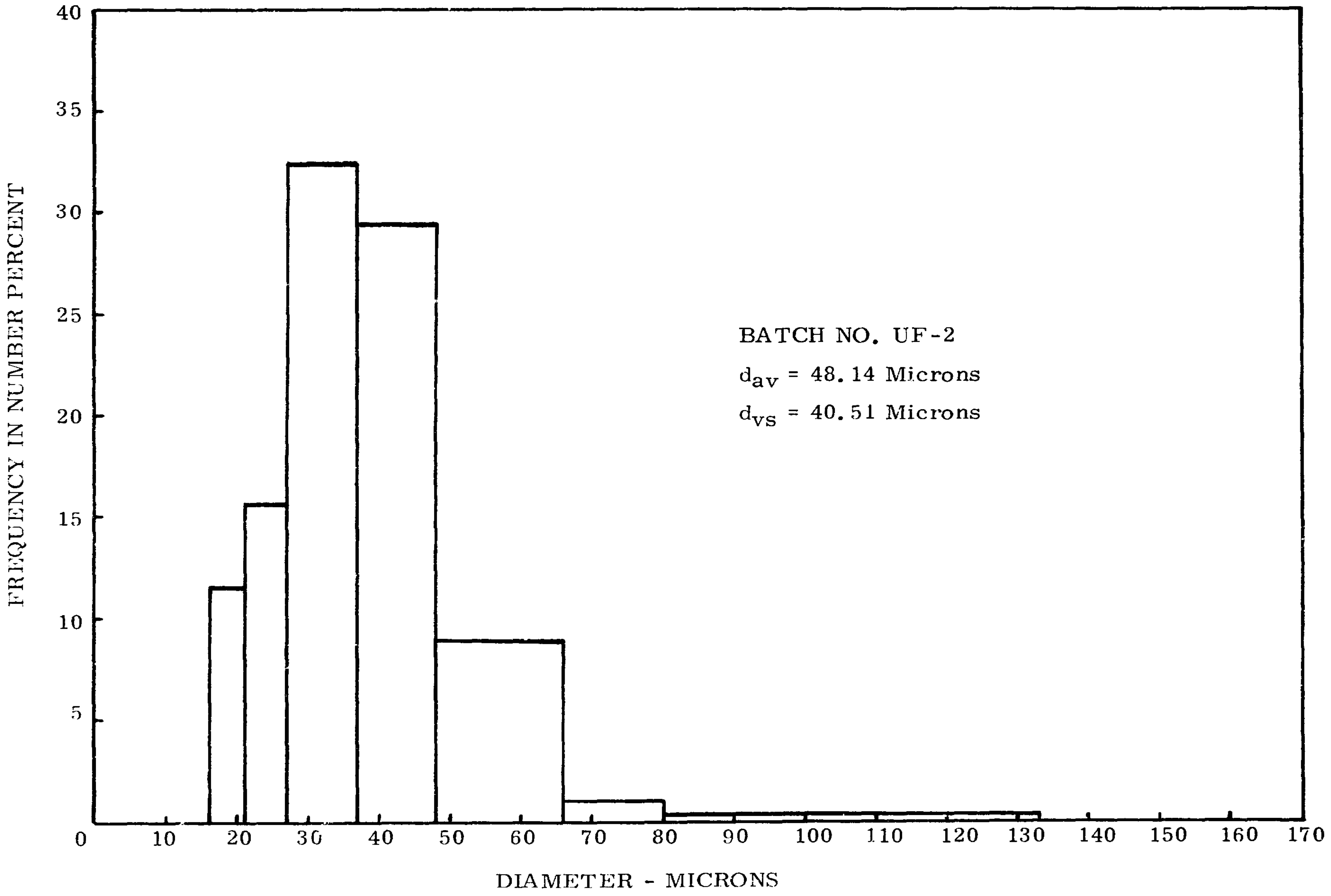
BATCH NO. UF-2
d_{av} = 48.14 Microns
d_{vs} = 40.51 Microns
FREQUENCY IN NUMBER PERCENT
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10
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25
30
35
40
0
10
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30
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80
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100
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140
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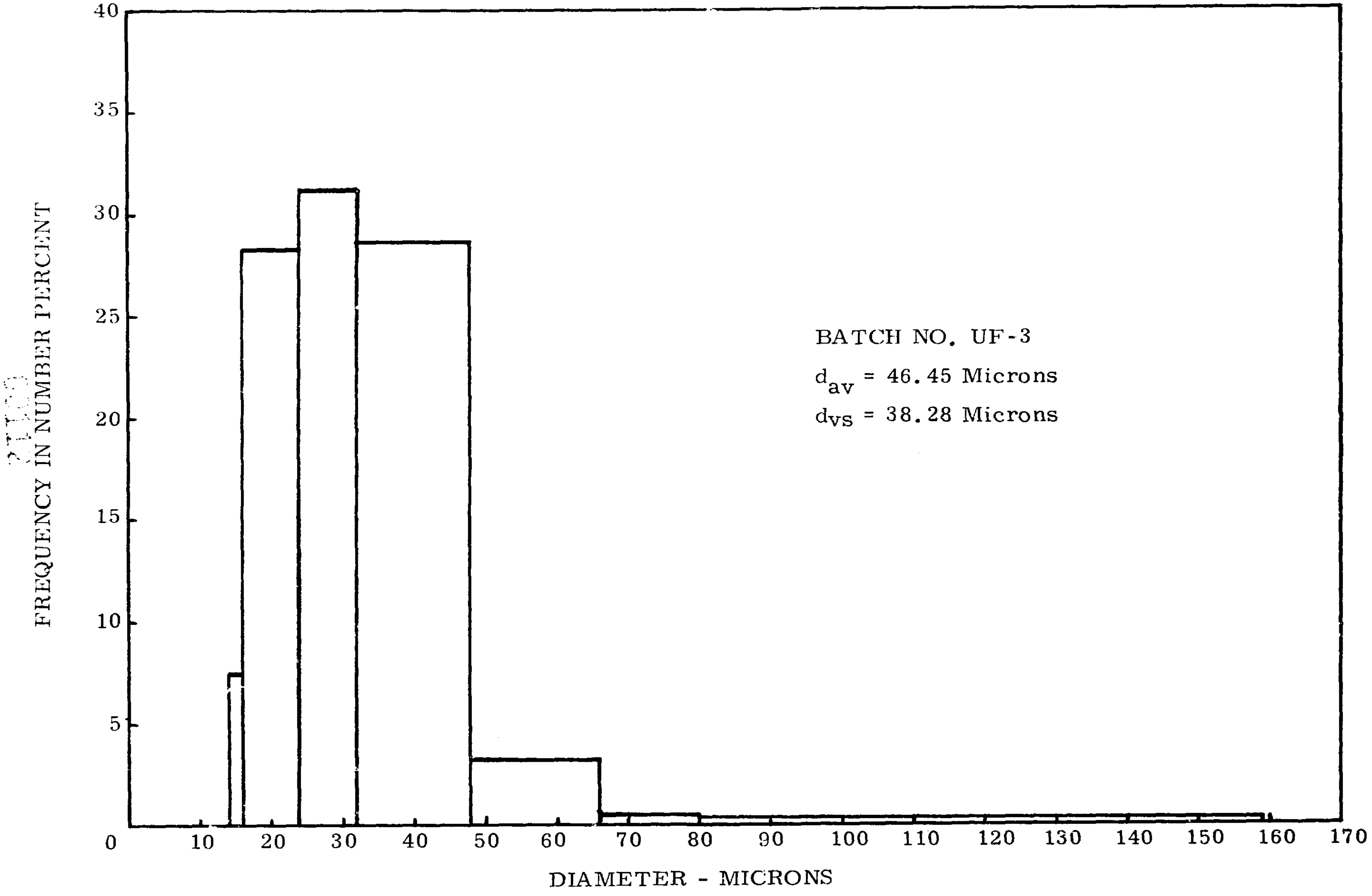
170
DIAMETER - MICRONS


BATCH NO. UF-3
d_{av} = 46.45 Microns
d_{vs} = 38.28 Microns
FREQUENCY IN NUMBER PERCENT
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35
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80
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100
110
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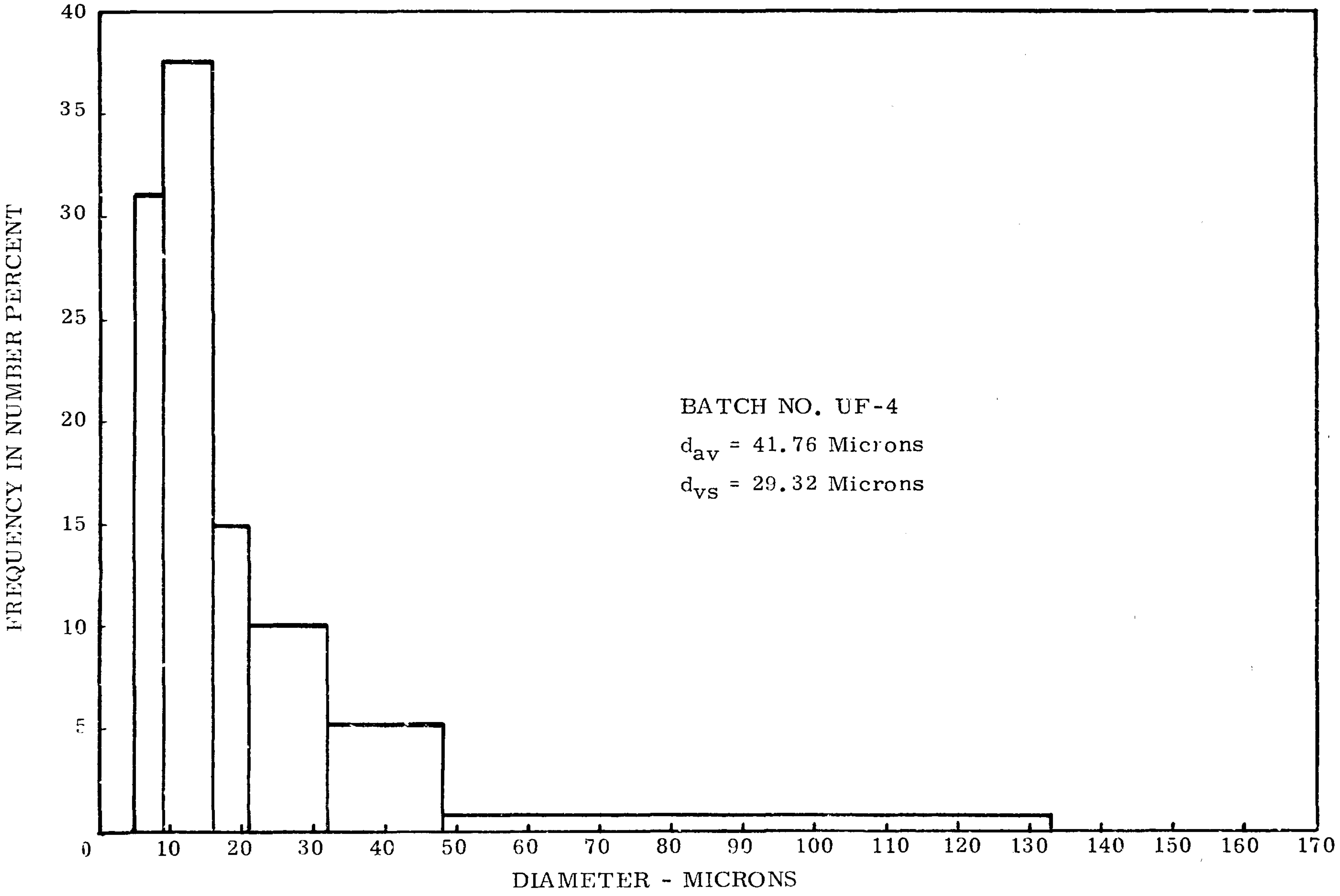
130
140
150
160
170
DIAMETER - MICRONS


BATCH NO. UF-4
d_{av} = 41.76 Microns
d_{vs} = 29.32 Microns
FREQUENCY IN NUMBER PERCENT
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35
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20
15
10
5
0
10
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80
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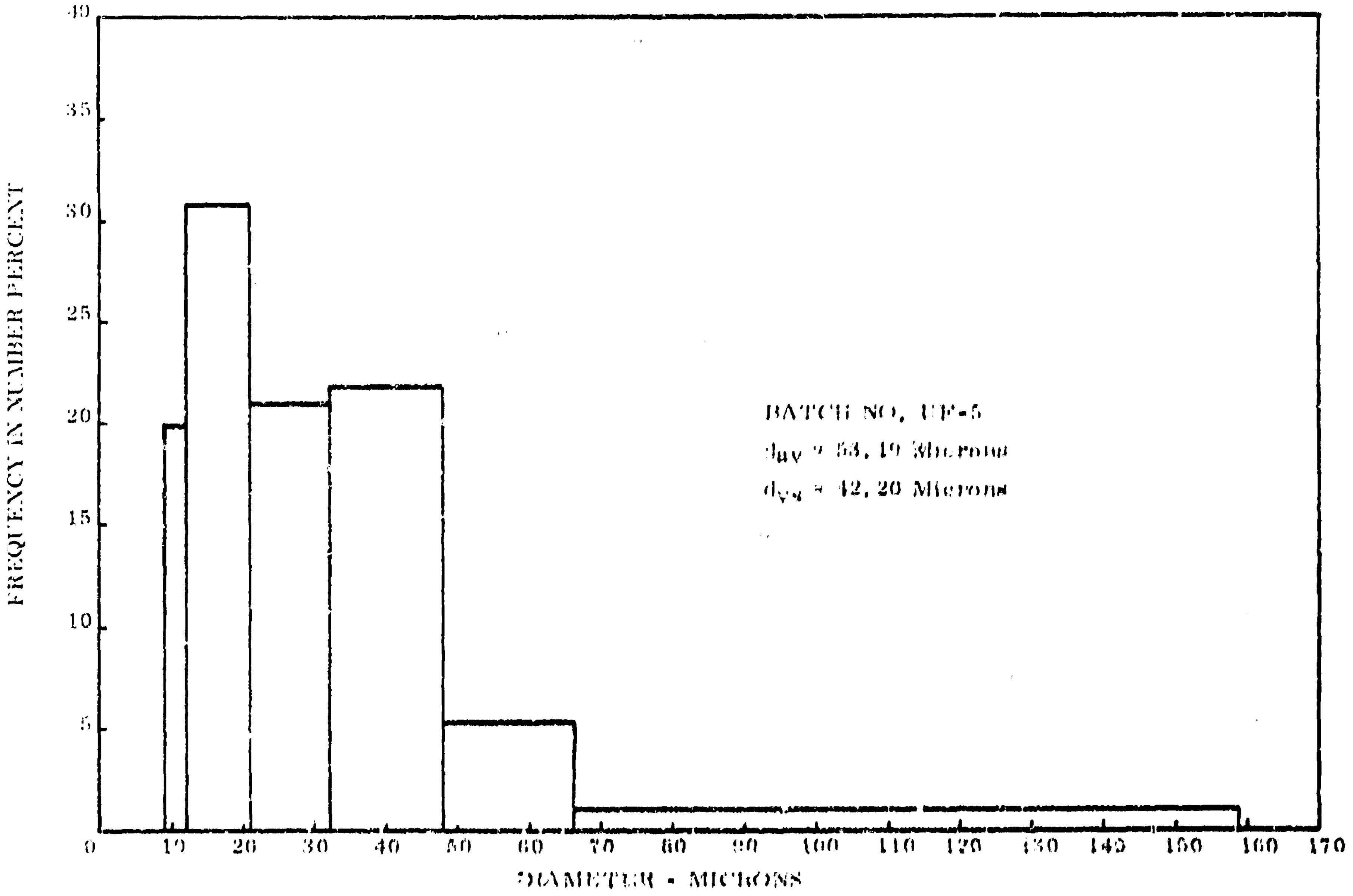
110
120
130
140
150
160
170
DIAMETER - MICRONS


FREQUENCY IN NUMBER PERCENT
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35
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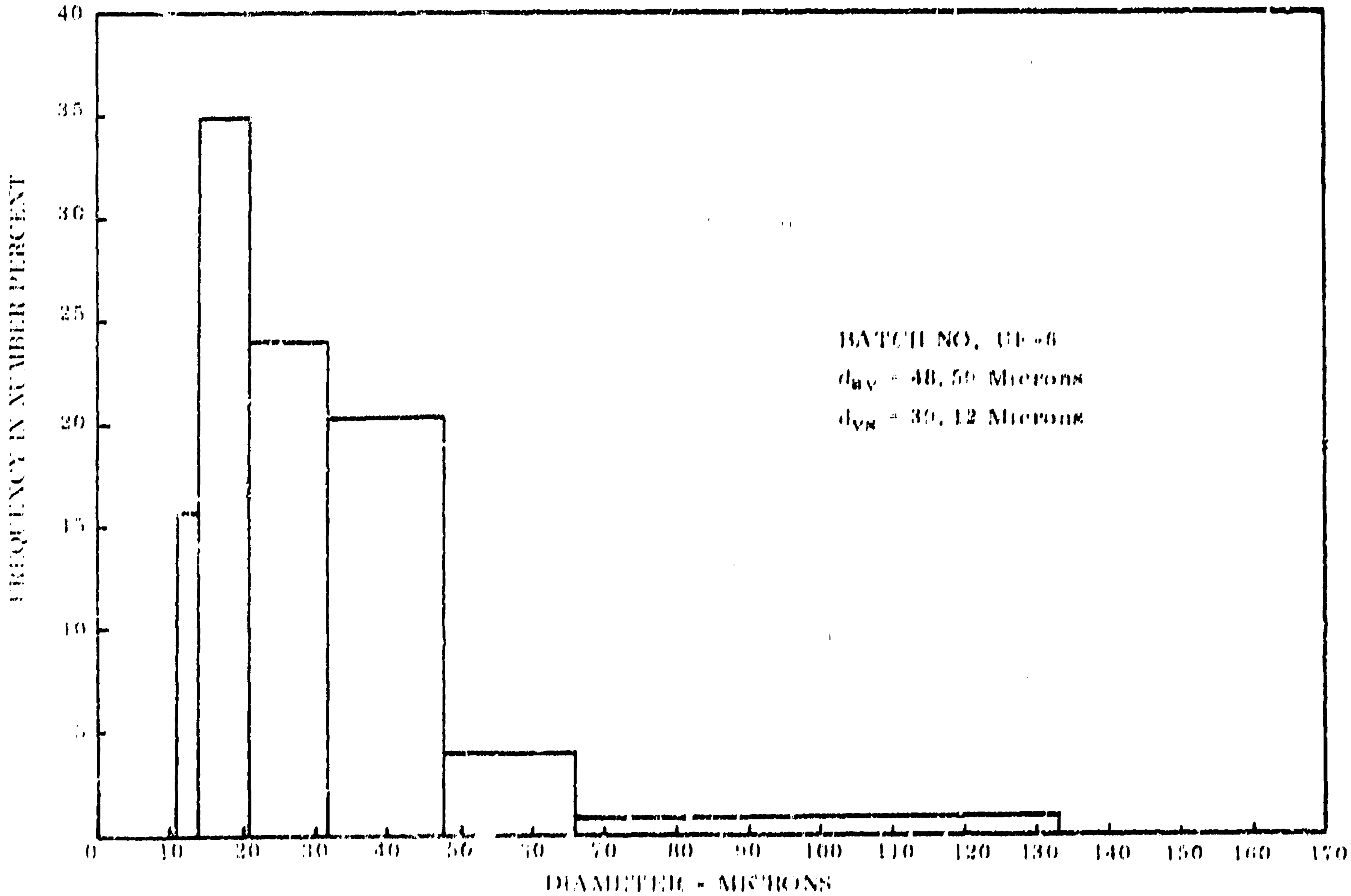
25
20
15
10
5
0
10
20
30
40
50
60
70
80
90
100
110
120
130
140
150
160
170
DIAMETER - MICRONS
BATCH NO. UF-5
d_{av} = 53.19 Microns
d_{vs} = 42.20 Microns


BATCH NO. UF-6
d_{av} = 48.50 Microns
d_{vs} = 30.12 Microns
FREQUENCY IN NUMBER PERCENT
DIAMETER - MICRONS
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5
10
15
20
25
30
35
40
0
10
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100
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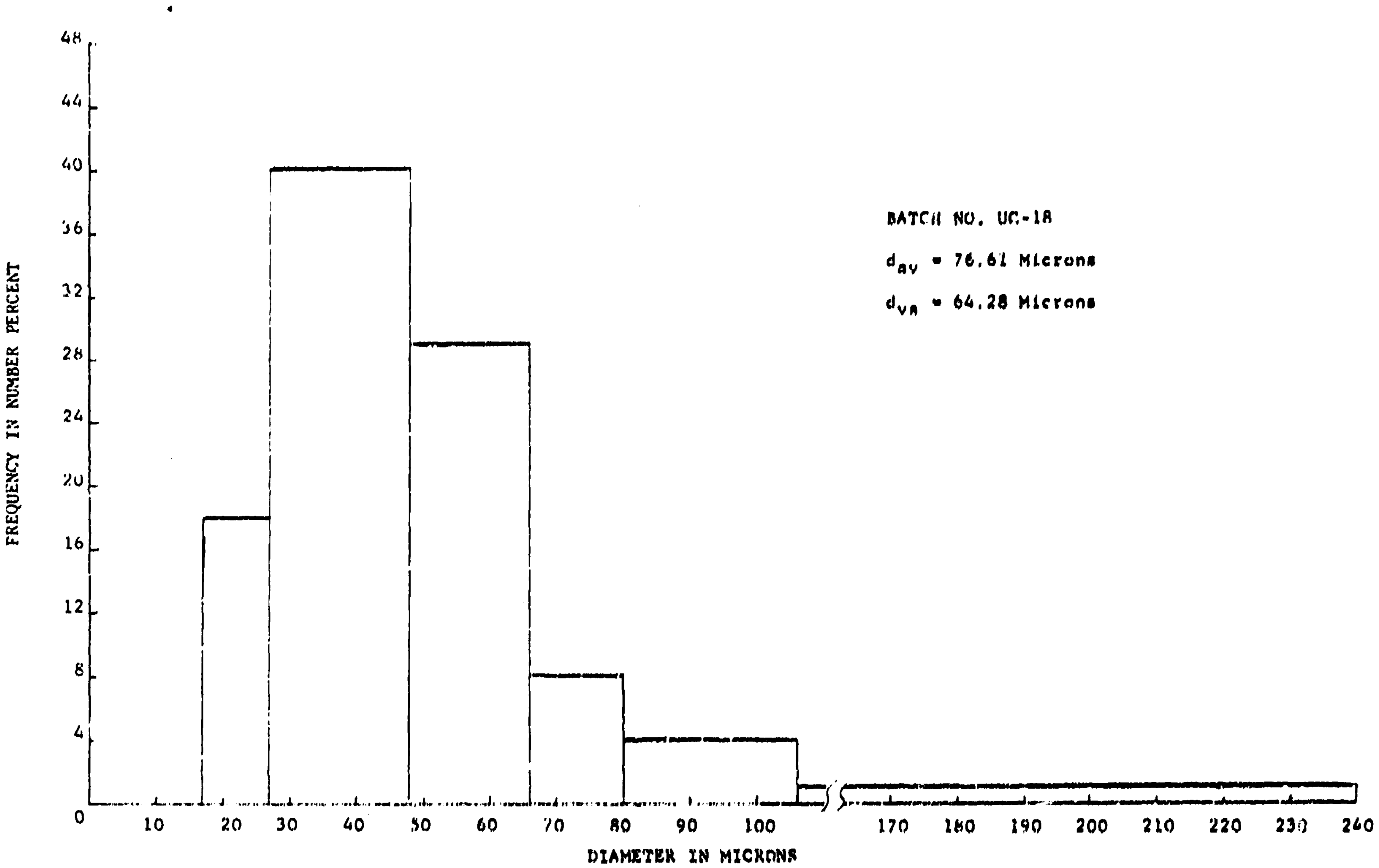
160
170


BATCH NO. UC-18
d_{av} = 76.61 Microns
d_{vs} = 64.28 Microns
FREQUENCY IN NUMBER PERCENT
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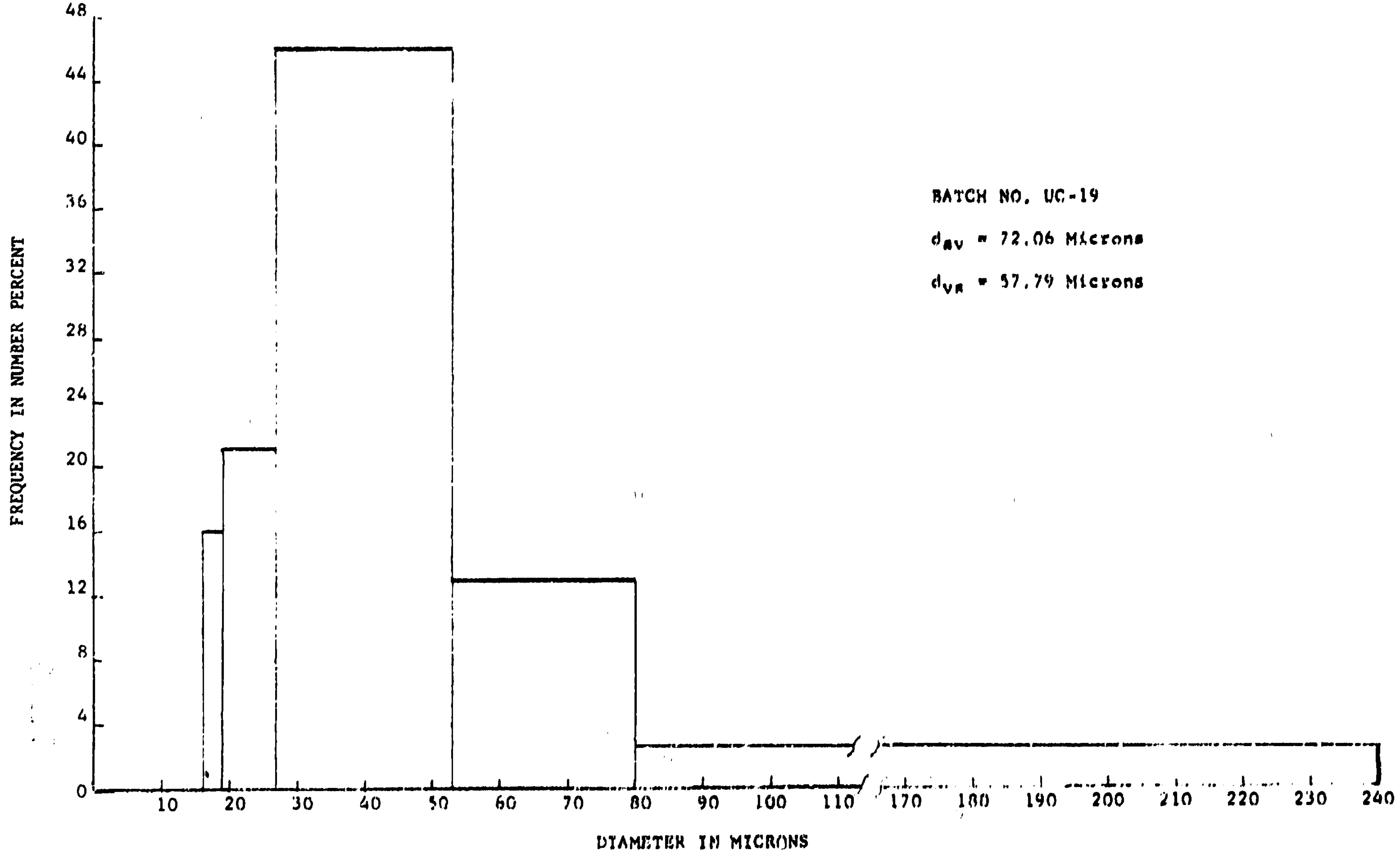
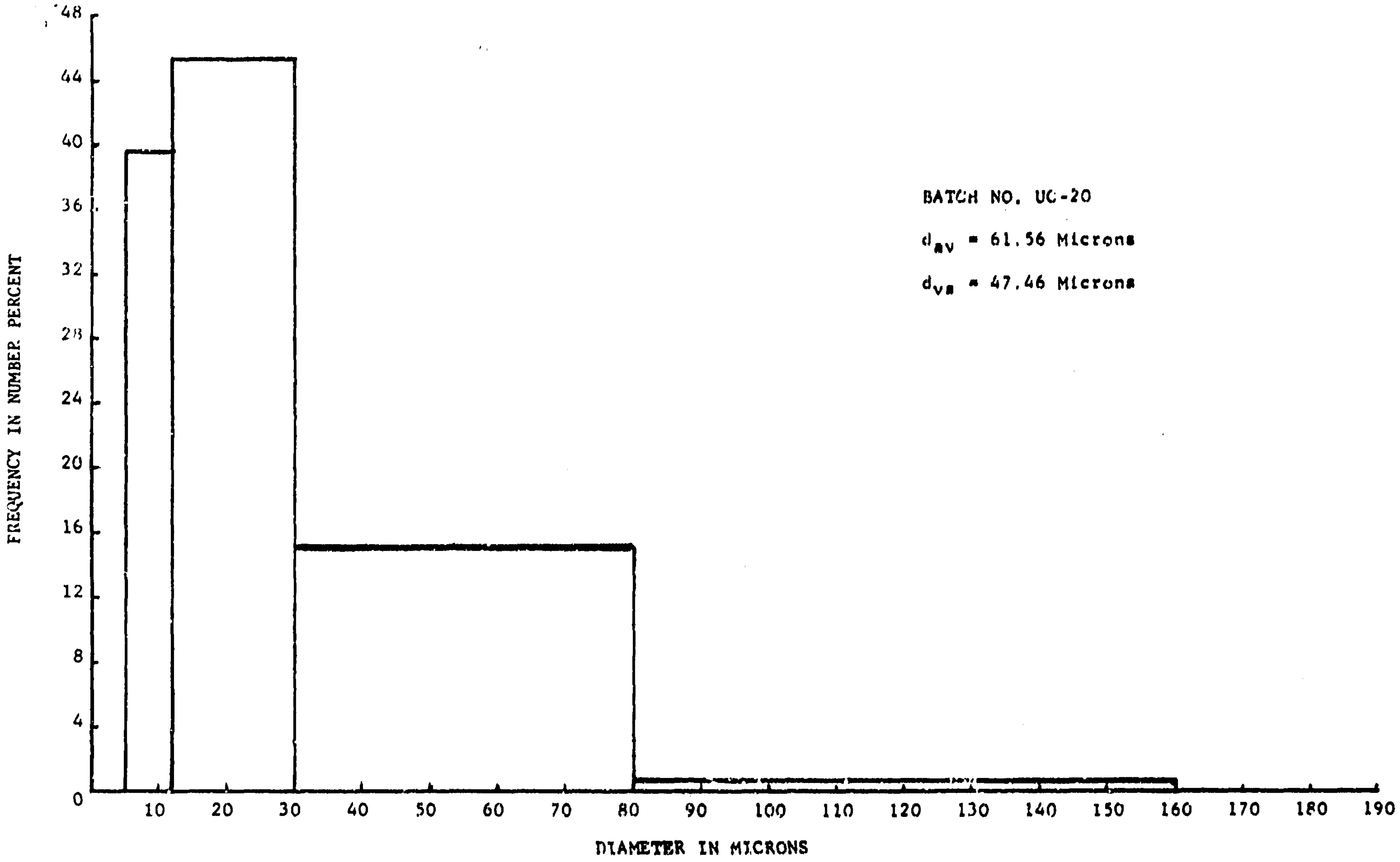
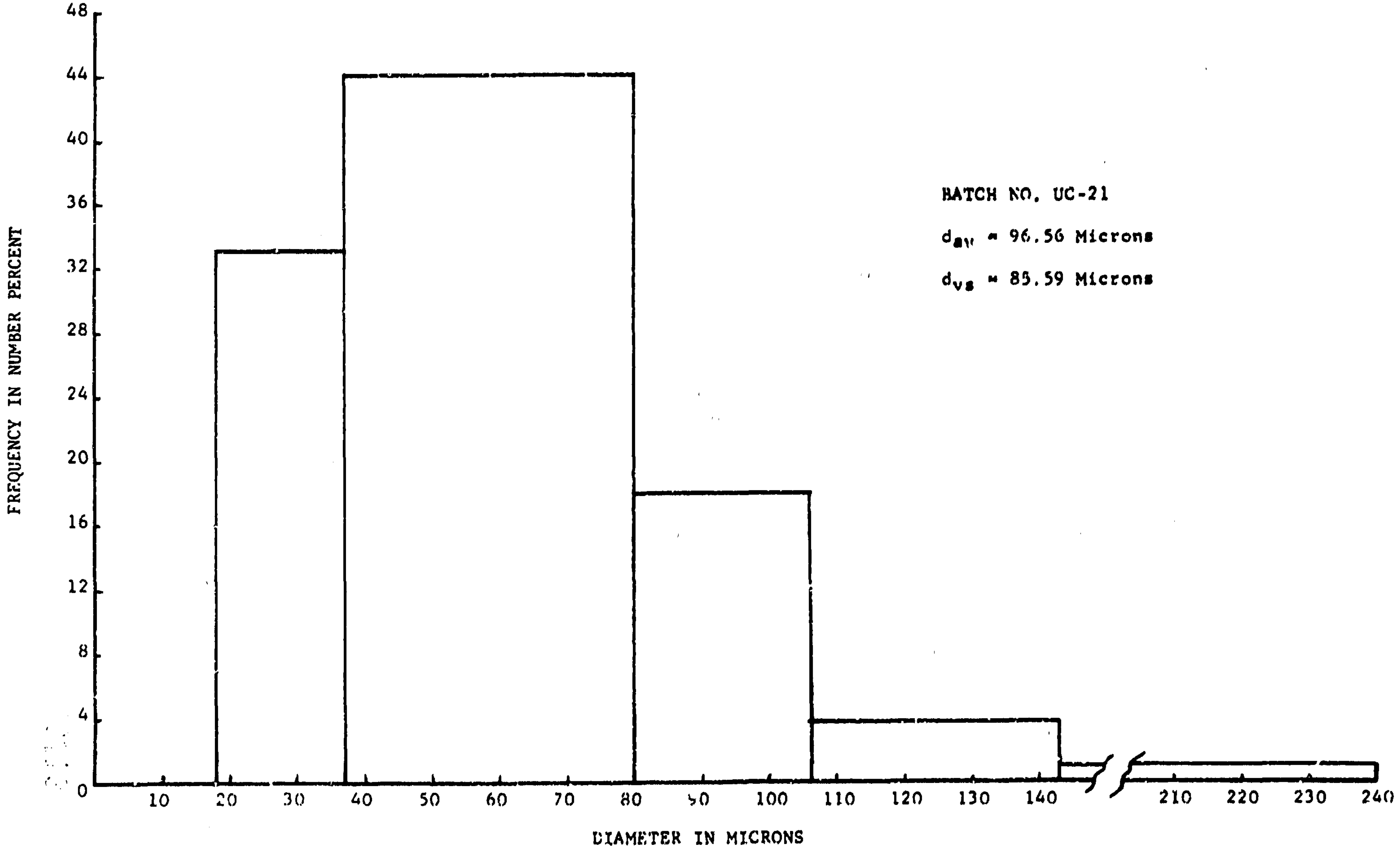
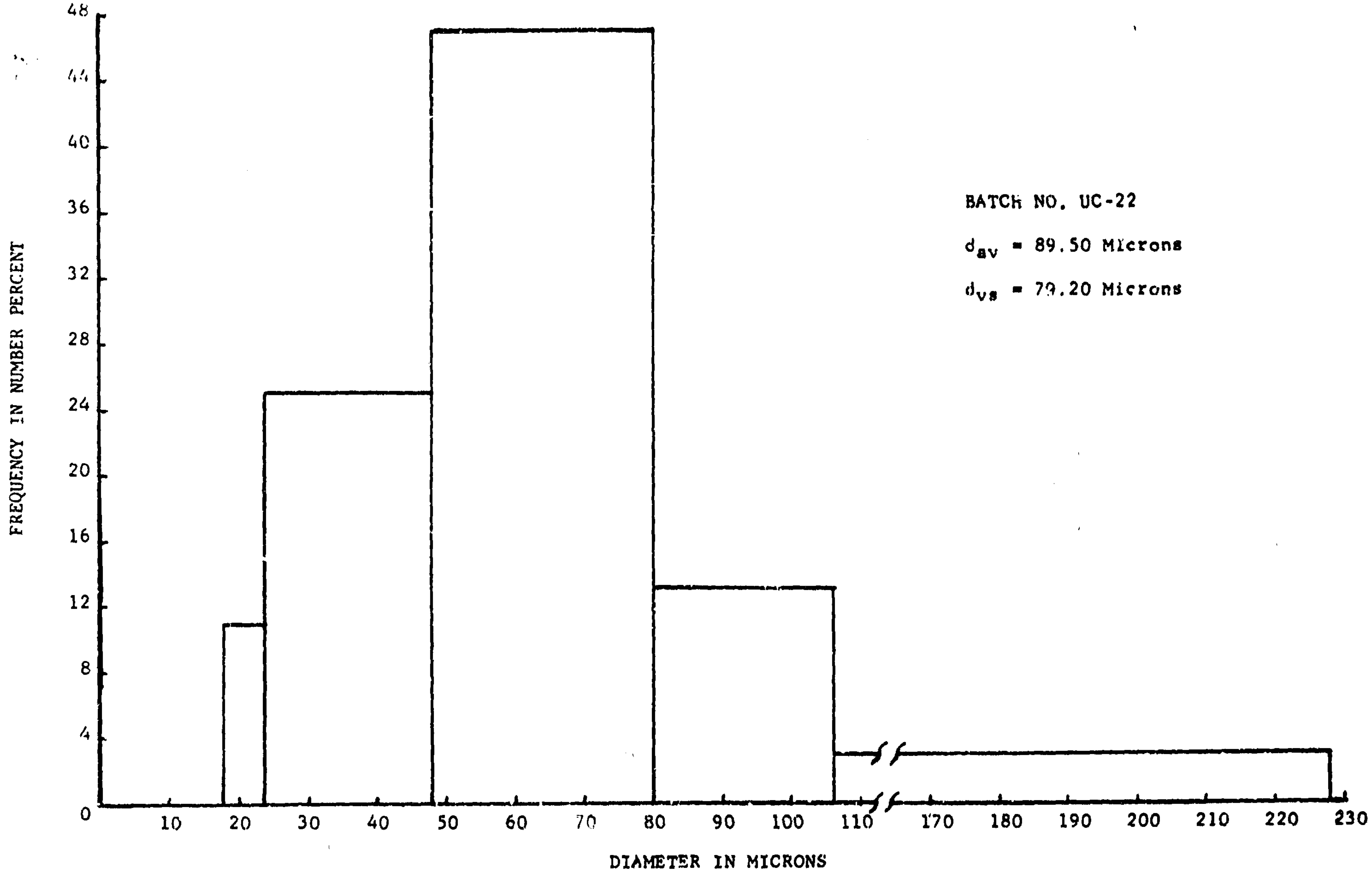
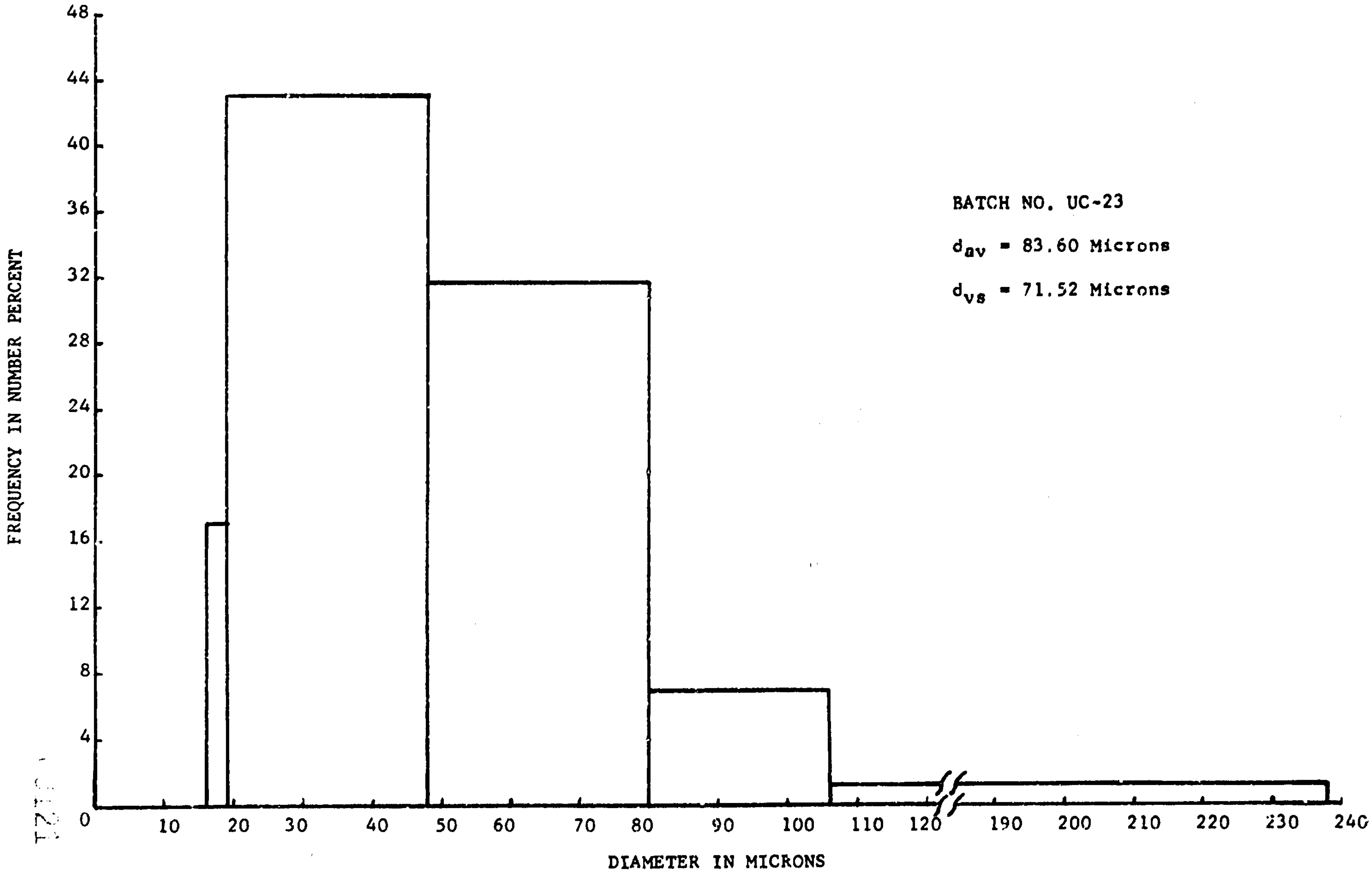
4
8
12
16
20
24
28
32
36
40
44
48
10
20
30
40
50
60
70
80
90
100
170
180
190
200
210
220
230
240
DIAMETER IN MICRONS

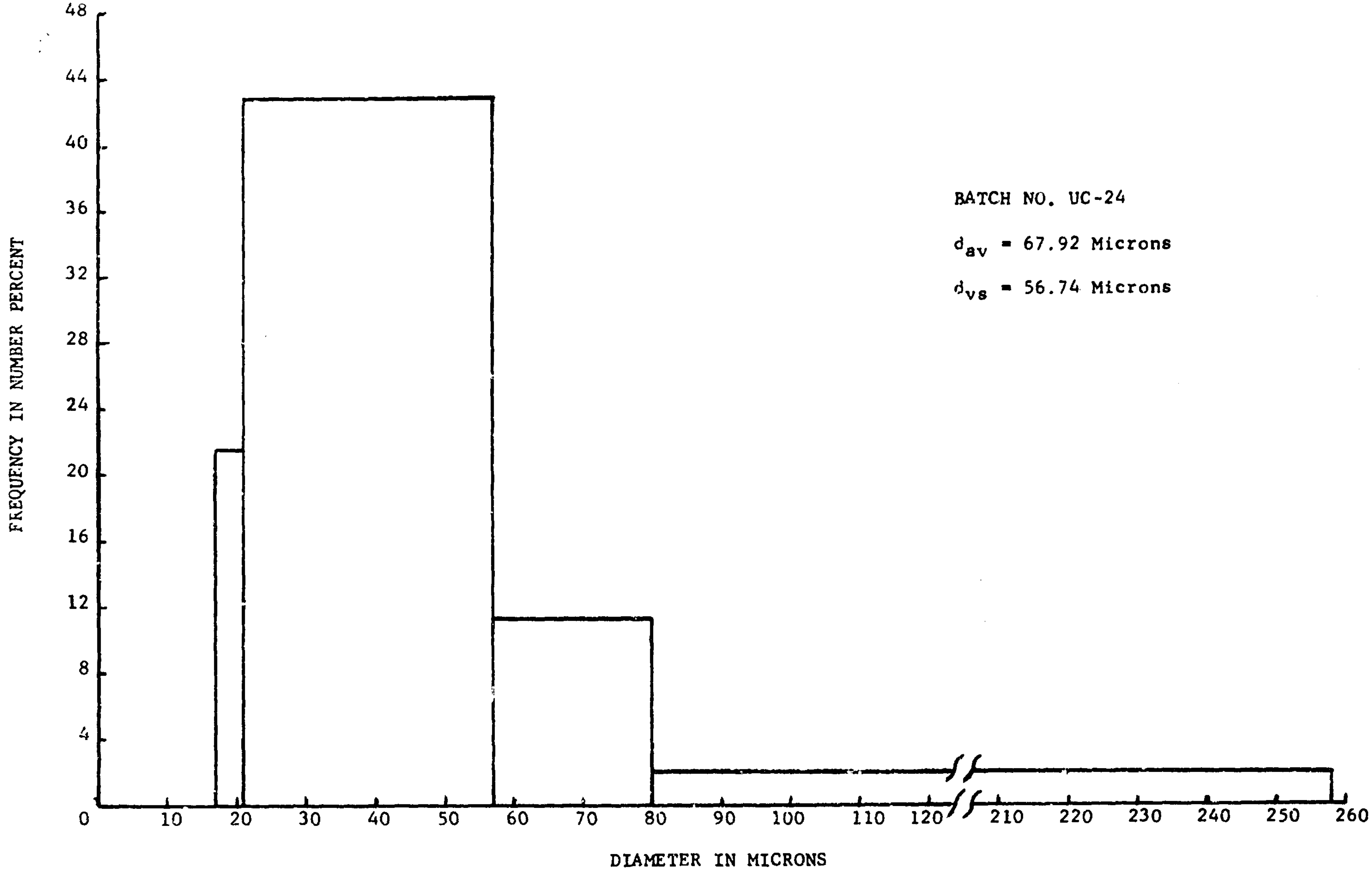

BATCH NO. UC-19
d_{av} = 72.06 Microns
d_{vs} = 57.79 Microns
FREQUENCY IN NUMBER PERCENT
0
4
8
12
16
20
24
28
32
36
40
44
48
10
20
30
40
50
60
70
80
90
100
110
170
180
190
200
210
220
230
240
DIAMETER IN MICRONS

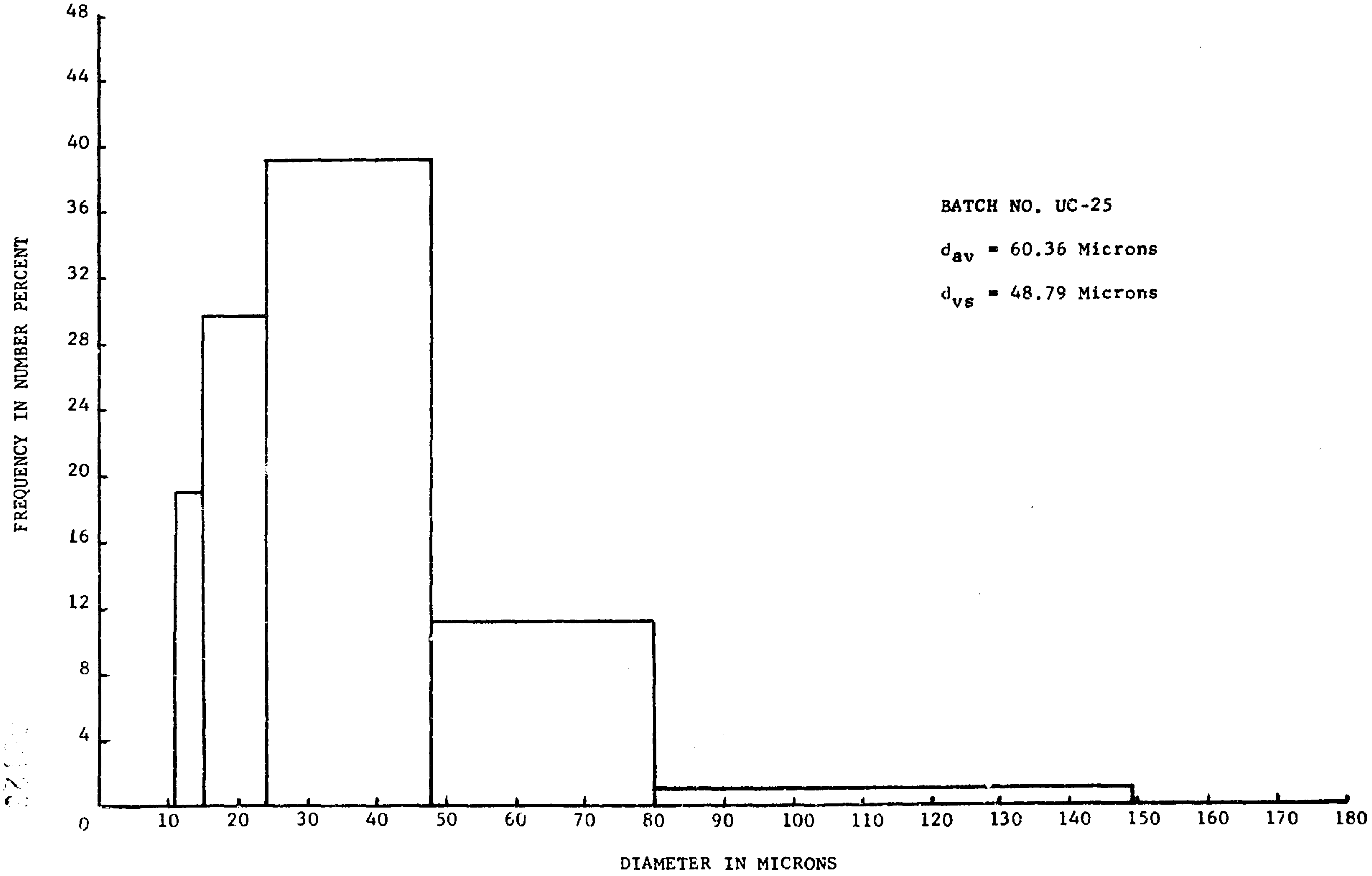

BATCH NO. UC-20
d_{av} = 61.56 Microns
d_{vs} = 47.46 Microns
FREQUENCY IN NUMBER PERCENT
0
4
8
12
16
20
24
28
32
36
40
44
48
DIAMETER IN MICRONS
10
20
30
40
50
60
70
80
90
100
110
120
130
140
150
160
170
180
190

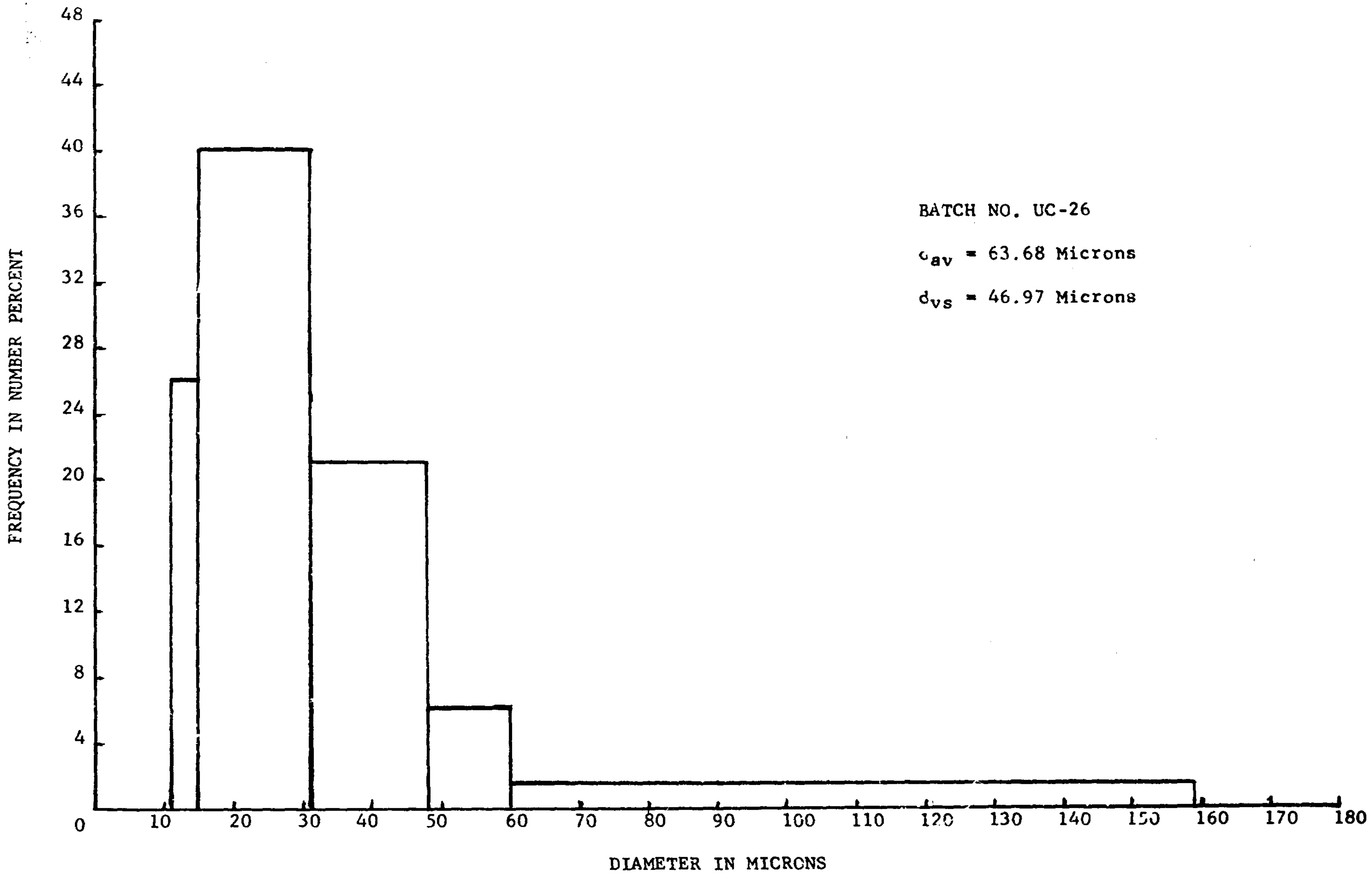

BATCH NO. UC-21
d_{av} = 96.56 Microns
d_{vs} = 85.59 Microns
FREQUENCY IN NUMBER PERCENT
48
44
40
36
32
28
24
20
16
12
8
4
0
10
20
30
40
50
60
70
80
90
100
110
120
130
140
210
220
230
240
DIAMETER IN MICRONS

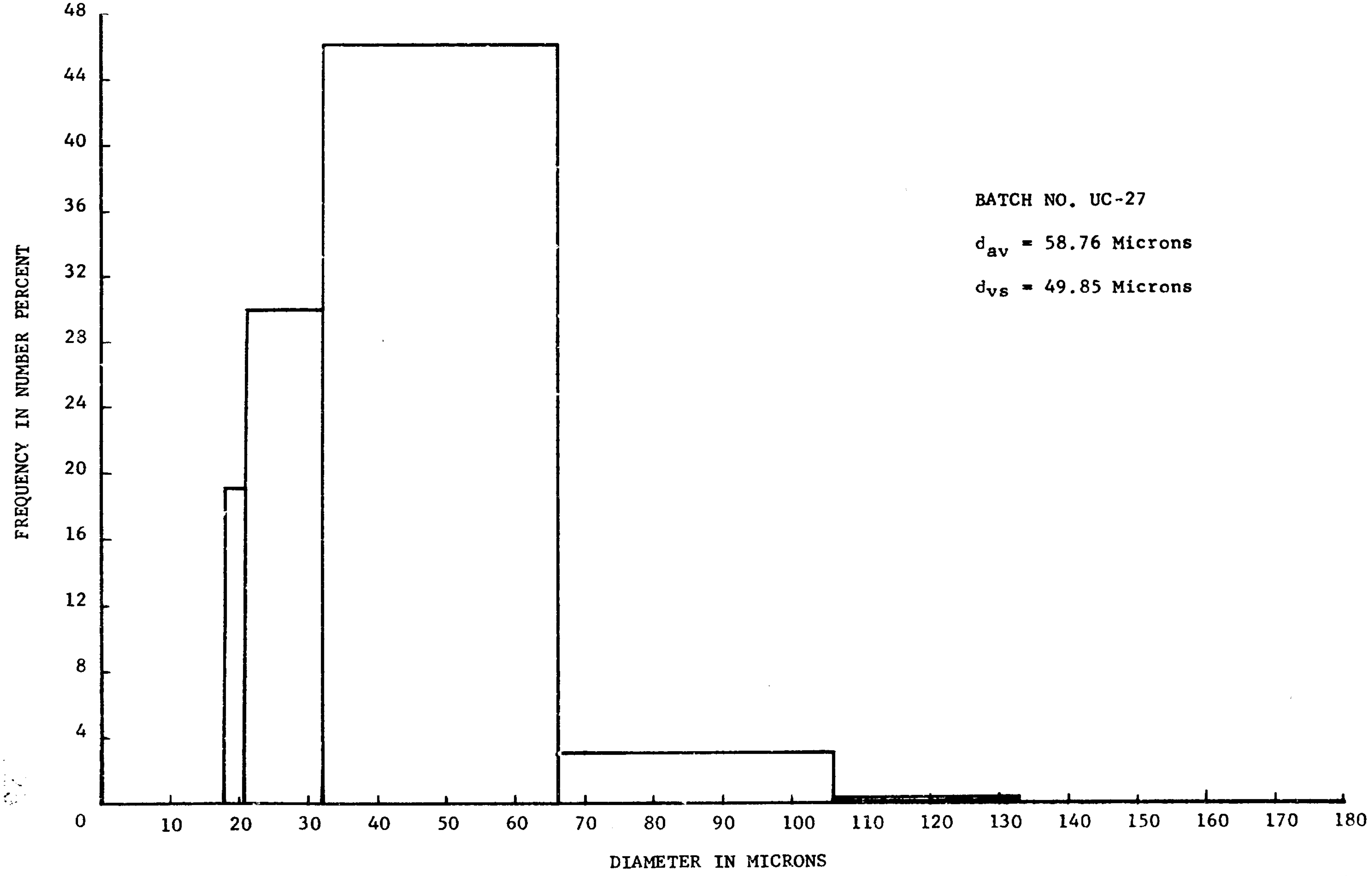

BATCH NO. UC-22
d_{av} = 89.50 Microns
d_{vs} = 79.20 Microns
FREQUENCY IN NUMBER PERCENT
0
4
8
12
16
20
24
28
32
36
40
44
48
10
20
30
40
50
60
70
80
90
100
110
170
180
190
200
210
220
230
DIAMETER IN MICRONS

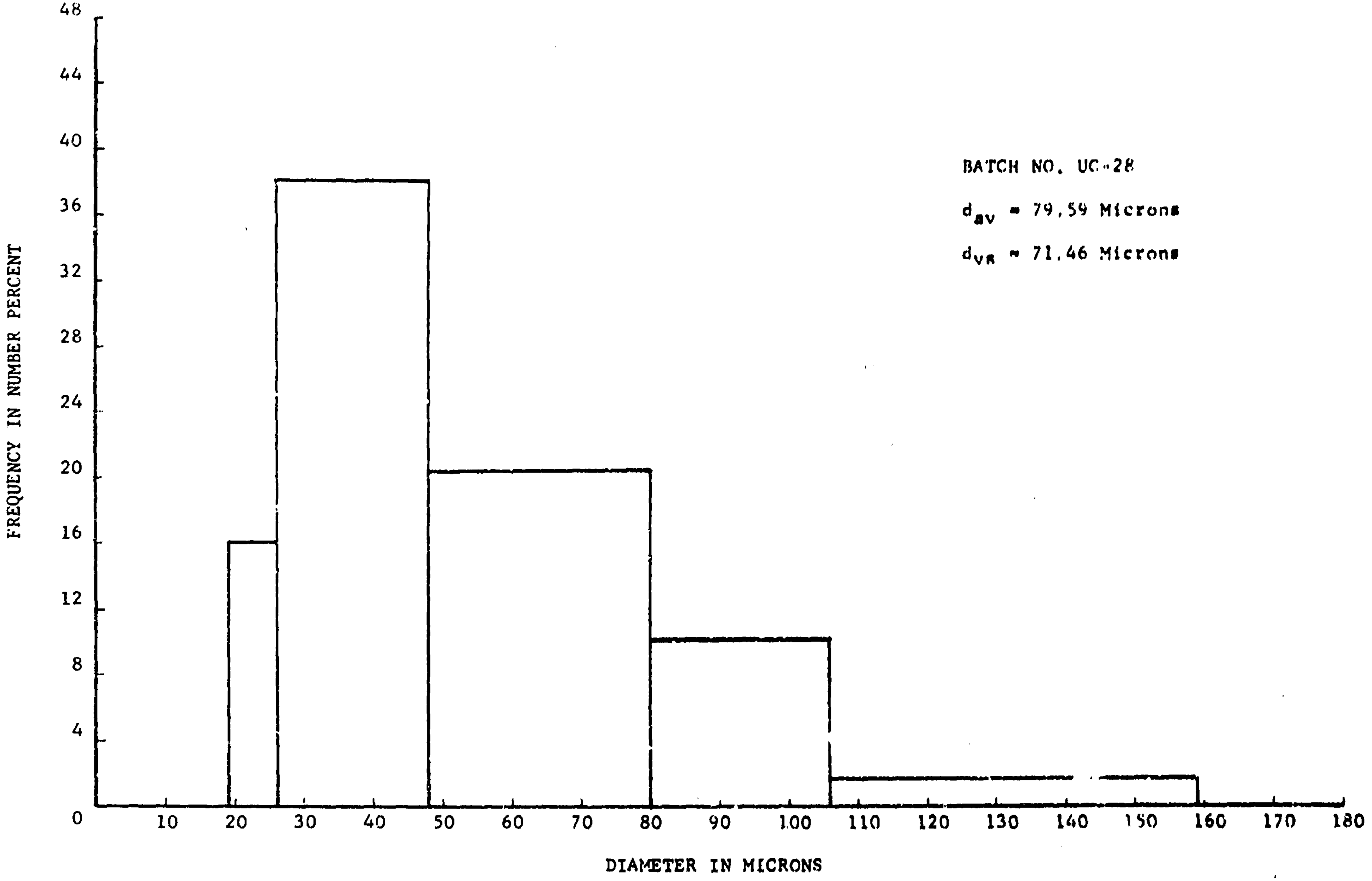

BATCH NO. UC-23
d_{av} = 83.60 Microns
d_{vs} = 71.52 Microns
FREQUENCY IN NUMBER PERCENT
48
44
40
36
32
28
24
20
16
12
8
4
0
10
20
30
40
50
60
70
80
90
100
110
120
190
200
210
220
230
240
DIAMETER IN MICRONS

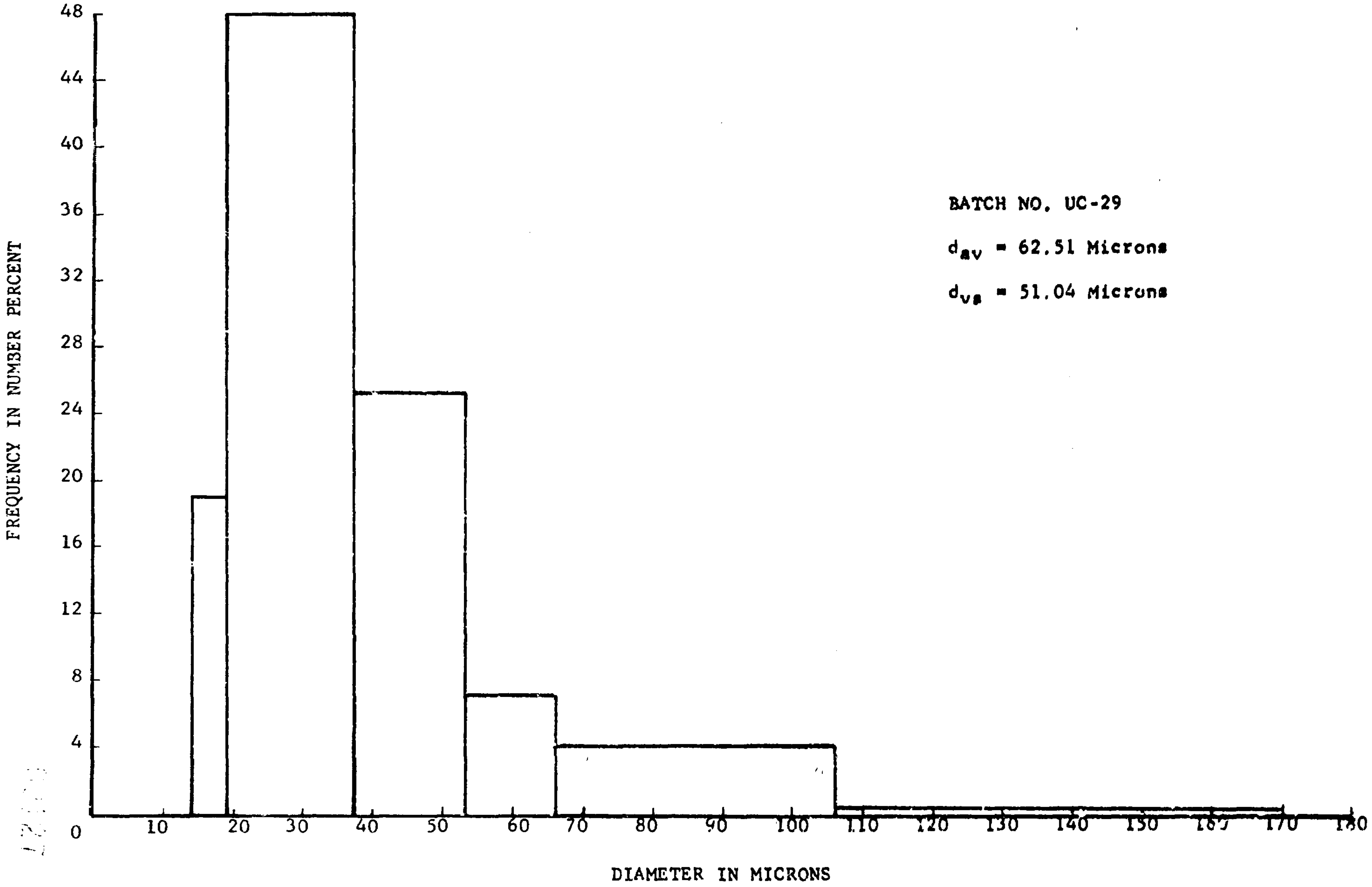

BATCH NO. UC-24
d_{av} = 67.92 Microns
d_{vs} = 56.74 Microns
FREQUENCY IN NUMBER PERCENT
0
4
8
12
16
20
24
28
32
36
40
44
48
DIAMETER IN MICRONS
10
20
30
40
50
60
70
80
90
100
110
120
210
220
230
240
250
260

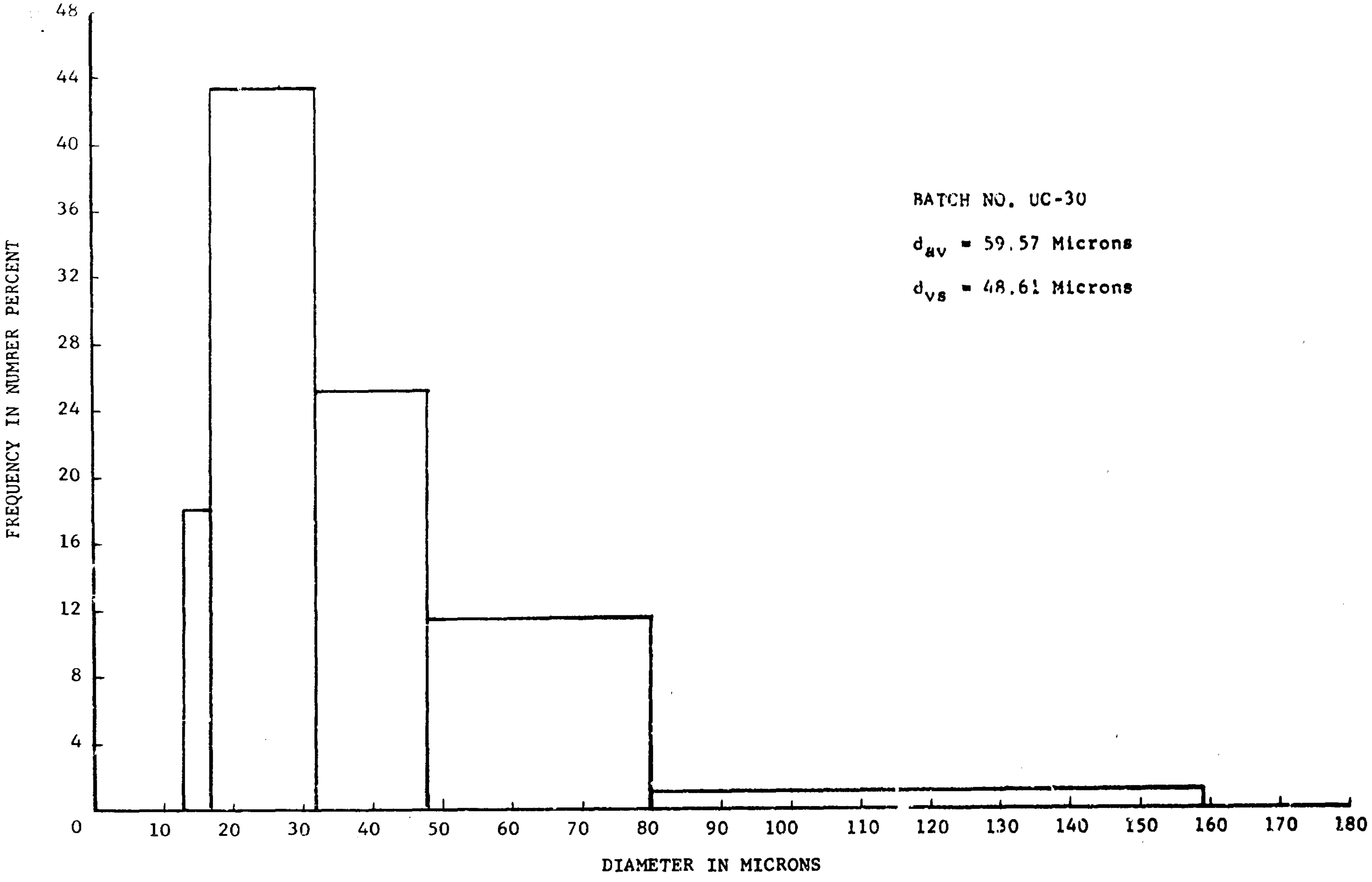

BATCH NO. UC-25
d_{av} = 60.36 Microns
d_{vs} = 48.79 Microns
FREQUENCY IN NUMBER PERCENT
0
4
8
12
16
20
24
28
32
36
40
44
48
10
20
30
40
50
60
70
80
90
100
110
120
130
140
150
160
170
180
DIAMETER IN MICRONS


BATCH NO. UC-26
d_{av} = 63.68 Microns
d_{vs} = 46.97 Microns
FREQUENCY IN NUMBER PERCENT
0
4
8
12
16
20
24
28
32
36
40
44
48
10
20
30
40
50
60
70
80
90
100
110
120
130
140
150
160
170
180
DIAMETER IN MICRONS


BATCH NO. UC-27
d_{av} = 58.76 Microns
d_{vs} = 49.85 Microns
FREQUENCY IN NUMBER PERCENT
48
44
40
36
32
28
24
20
16
12
8
4
0
10
20
30
40
50
60
70
80
90
100
110
120
130
140
150
160
170
180
DIAMETER IN MICRONS


BATCH NO. UC-28
d_av = 79,59 Microns
d_vR = 71.46 Microns
FREQUENCY IN NUMBER PERCENT
0
4
8
12
16
20
24
28
32
36
40
44
48
10
20
30
40
50
60
70
80
90
100
110
120
130
140
150
160
170
180
DIAMETER IN MICRONS

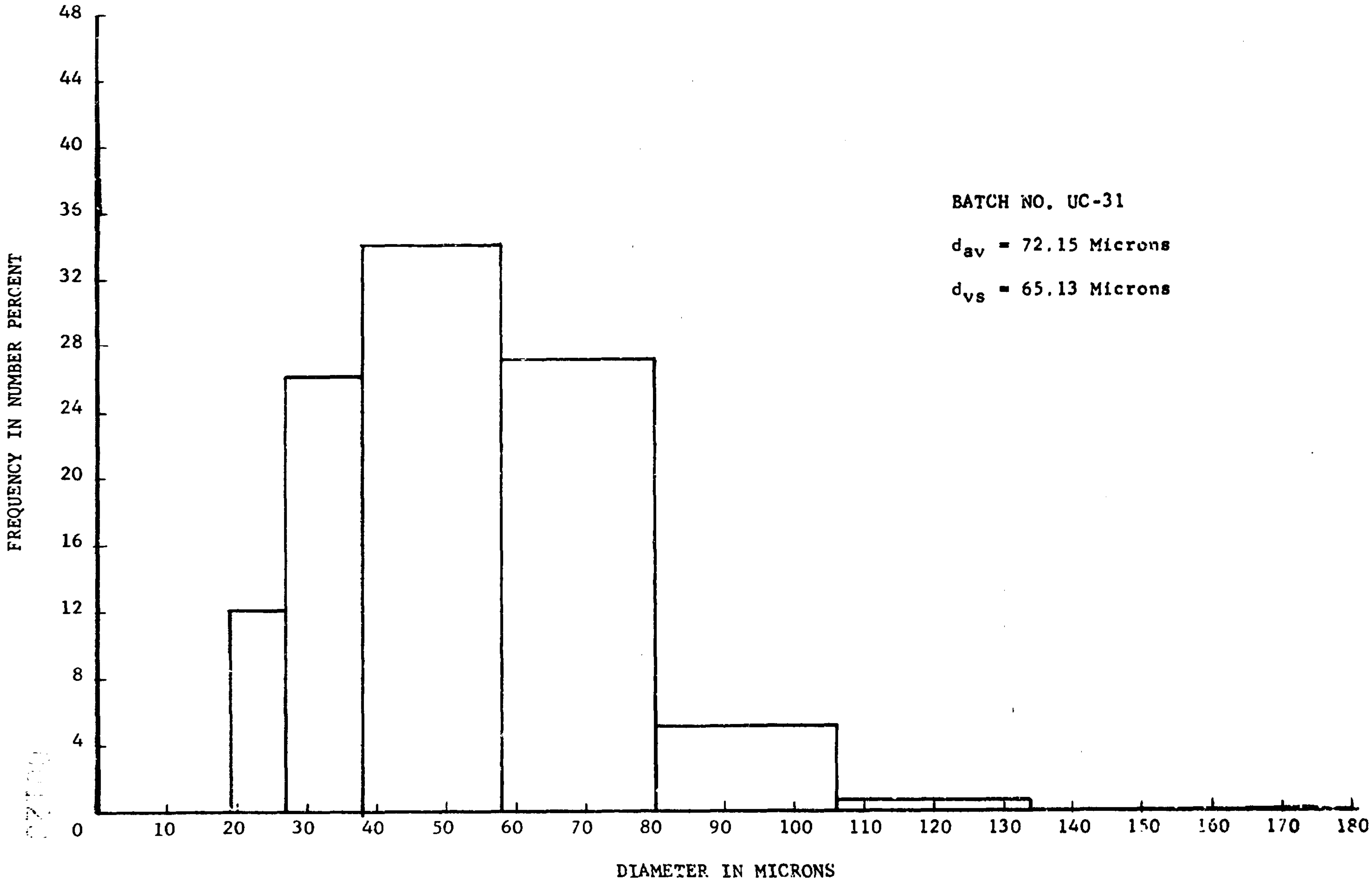

BATCH NO. UC-29
d_{av} = 62.51 Microns
d_{vs} = 51.04 Microns
FREQUENCY IN NUMBER PERCENT
0
4
8
12
16
20
24
28
32
36
40
44
48
10
20
30
40
50
60
70
80
90
100
110
120
130
140
150
160
170
180
DIAMETER IN MICRONS

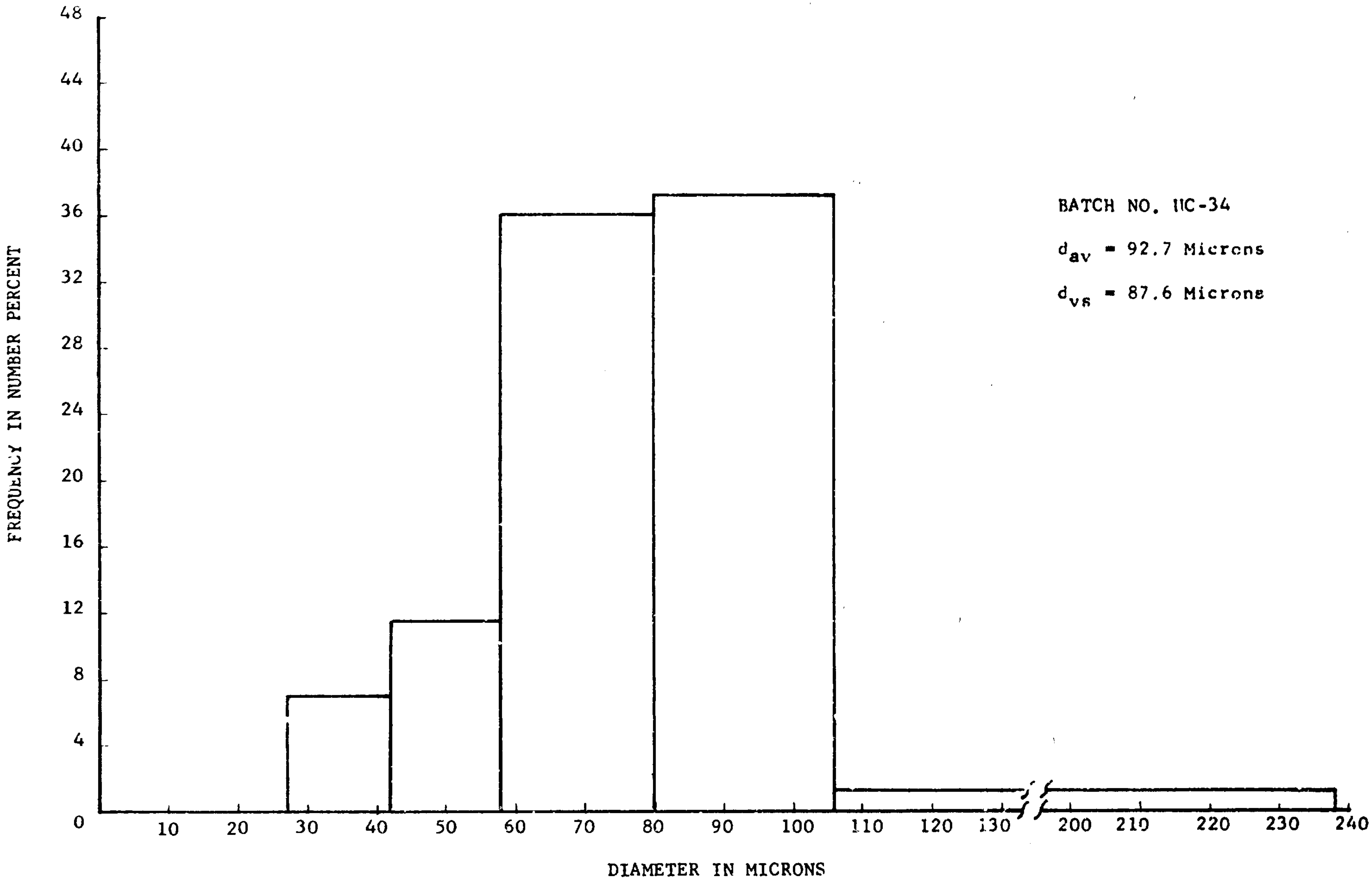


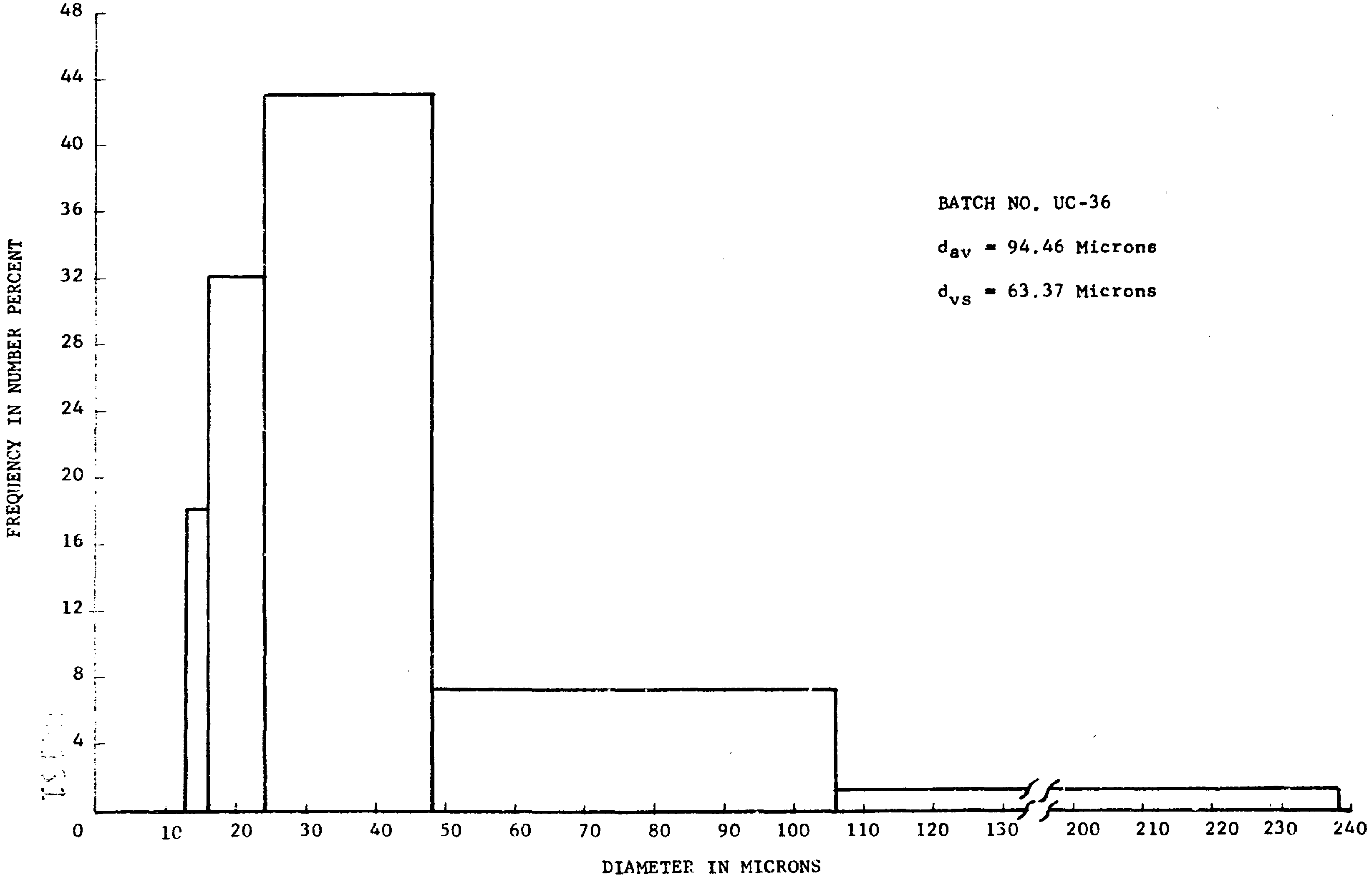
BATCH NO. UC-30

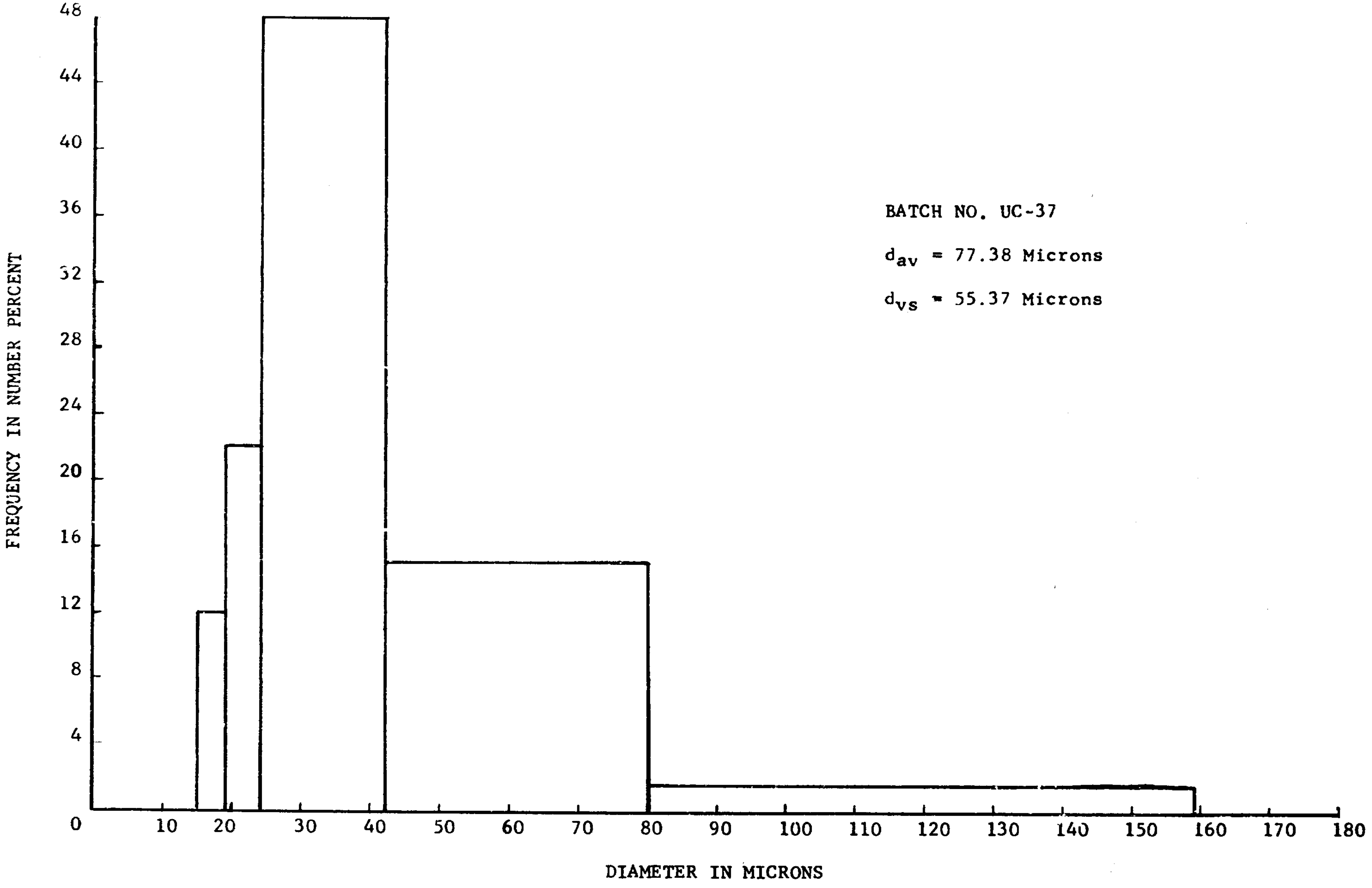
d_{av} = 59.57 Microns

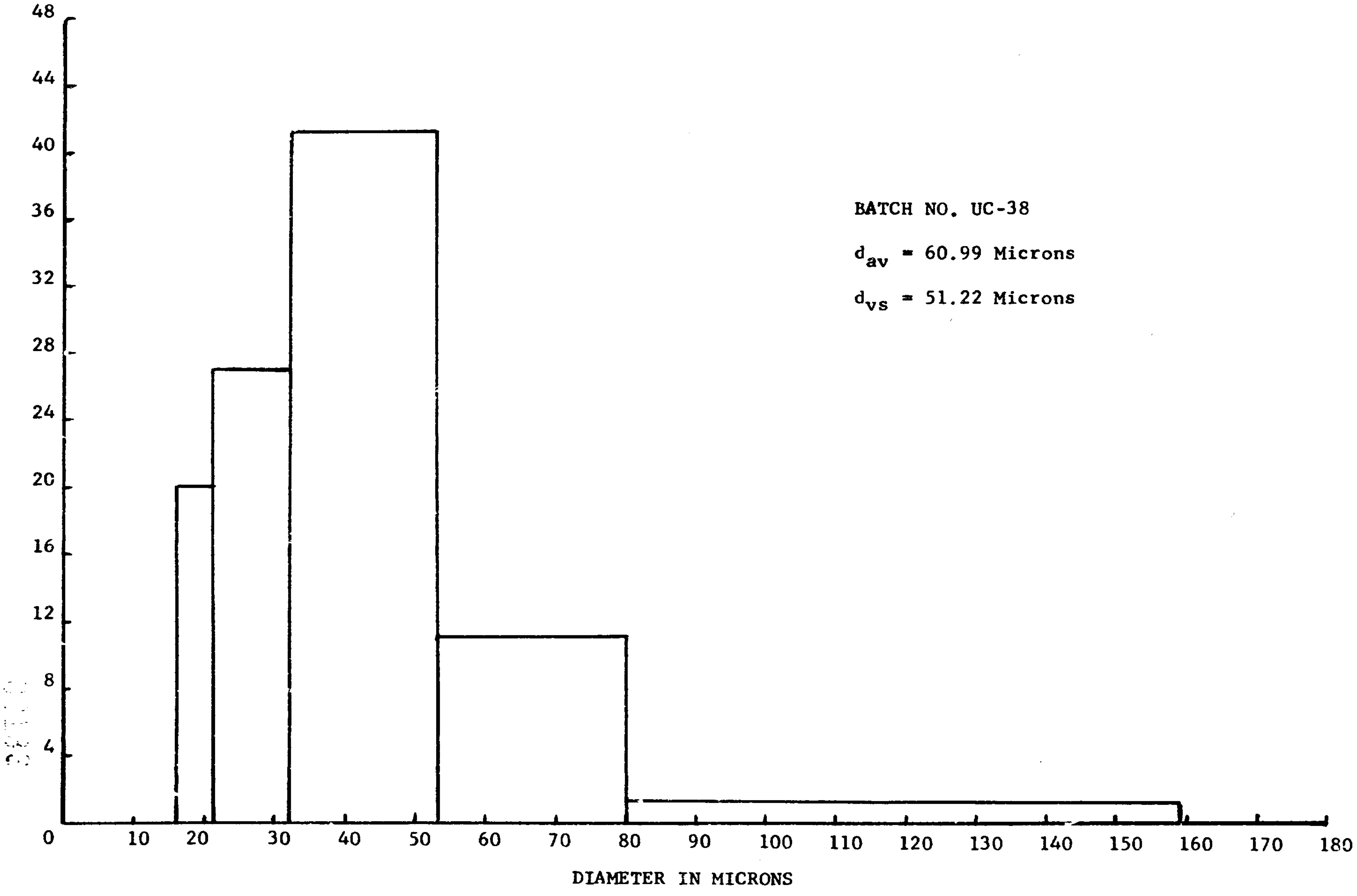
d_{vs} = 48.61 Microns

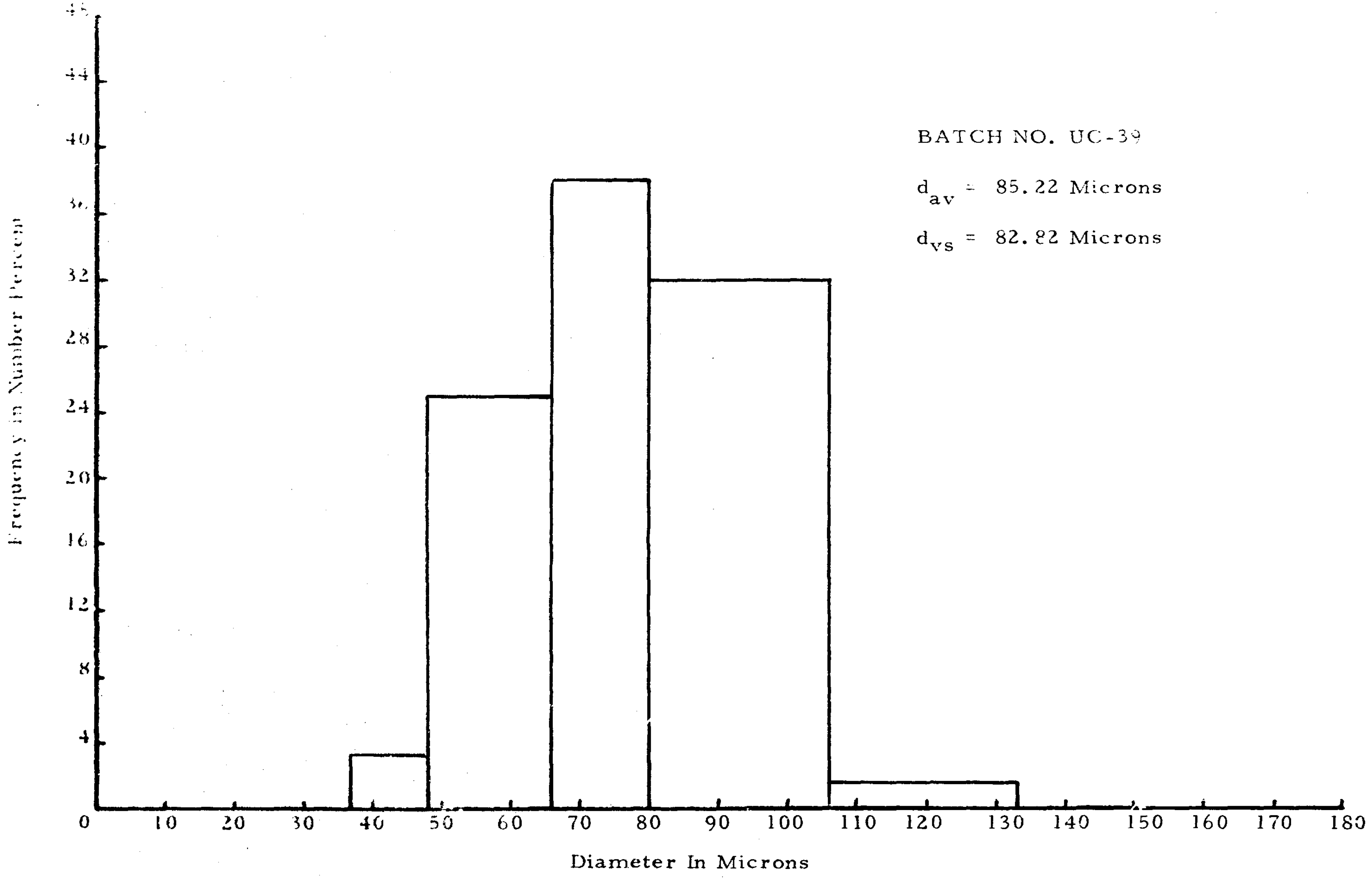

BATCH NO. UC-31
d_{av} = 72.15 Microns
d_{vs} = 65.13 Microns
FREQUENCY IN NUMBER PERCENT
0
4
8
12
16
20
24
28
32
36
40
44
48
10
20
30
40
50
60
70
80
90
100
110
120
130
140
150
160
170
180
DIAMETER IN MICRONS

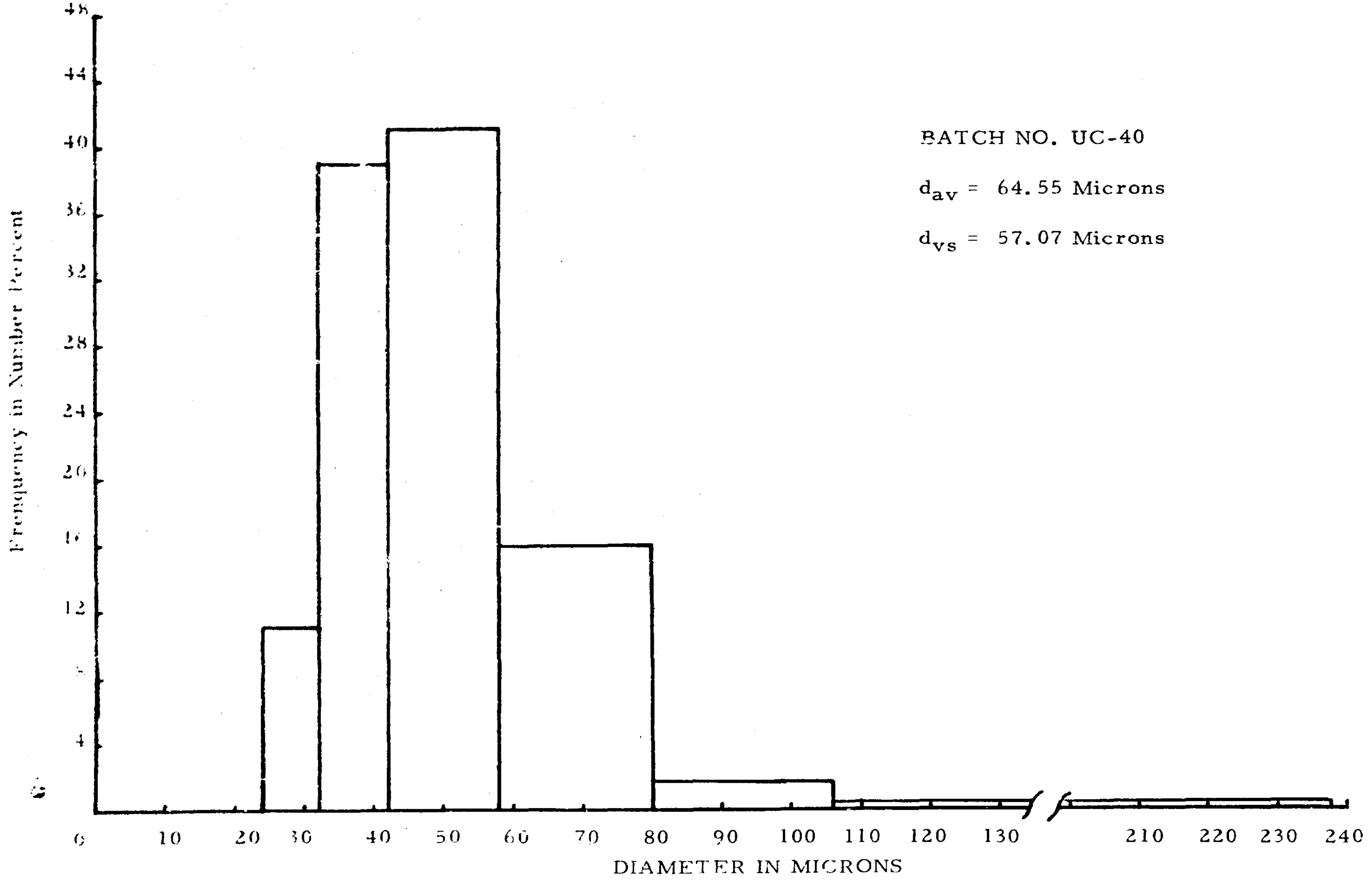

BATCH NO. HC-34
d_{av} = 92.7 Microns
d_{vs} = 87.6 Microns
FREQUENCY IN NUMBER PERCENT
48
44
40
36
32
28
24
20
16
12
8
4
0
10
20
30
40
50
60
70
80
90
100
110
120
130
200
210
220
230
240
DIAMETER IN MICRONS

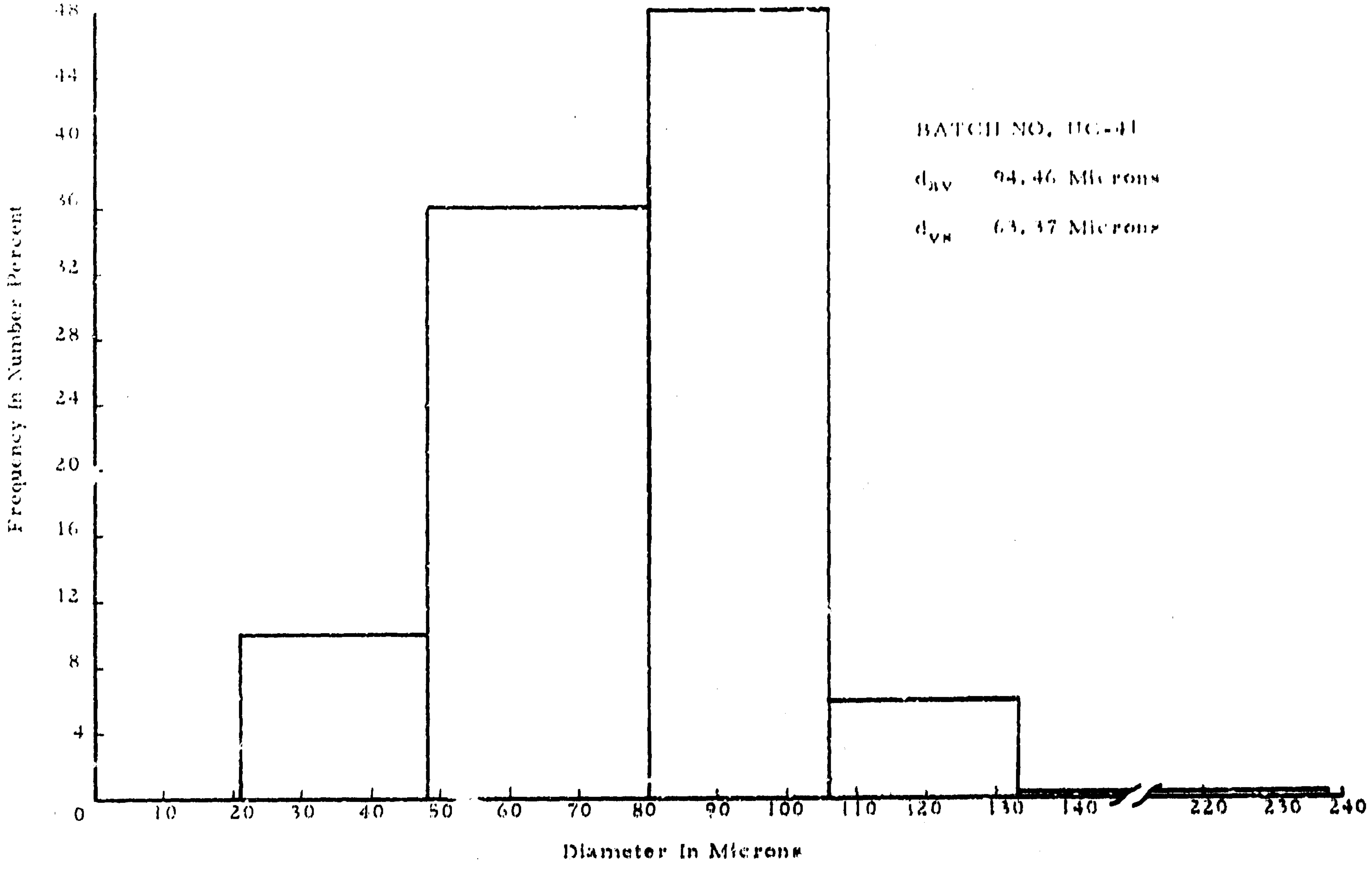

BATCH NO. UC-36
d_{av} = 94.46 Microns
d_{vs} = 63.37 Microns
FREQUENCY IN NUMBER PERCENT
0
4
8
12
16
20
24
28
32
36
40
44
48
10
20
30
40
50
60
70
80
90
100
110
120
130
200
210
220
230
240
DIAMETER IN MICRONS

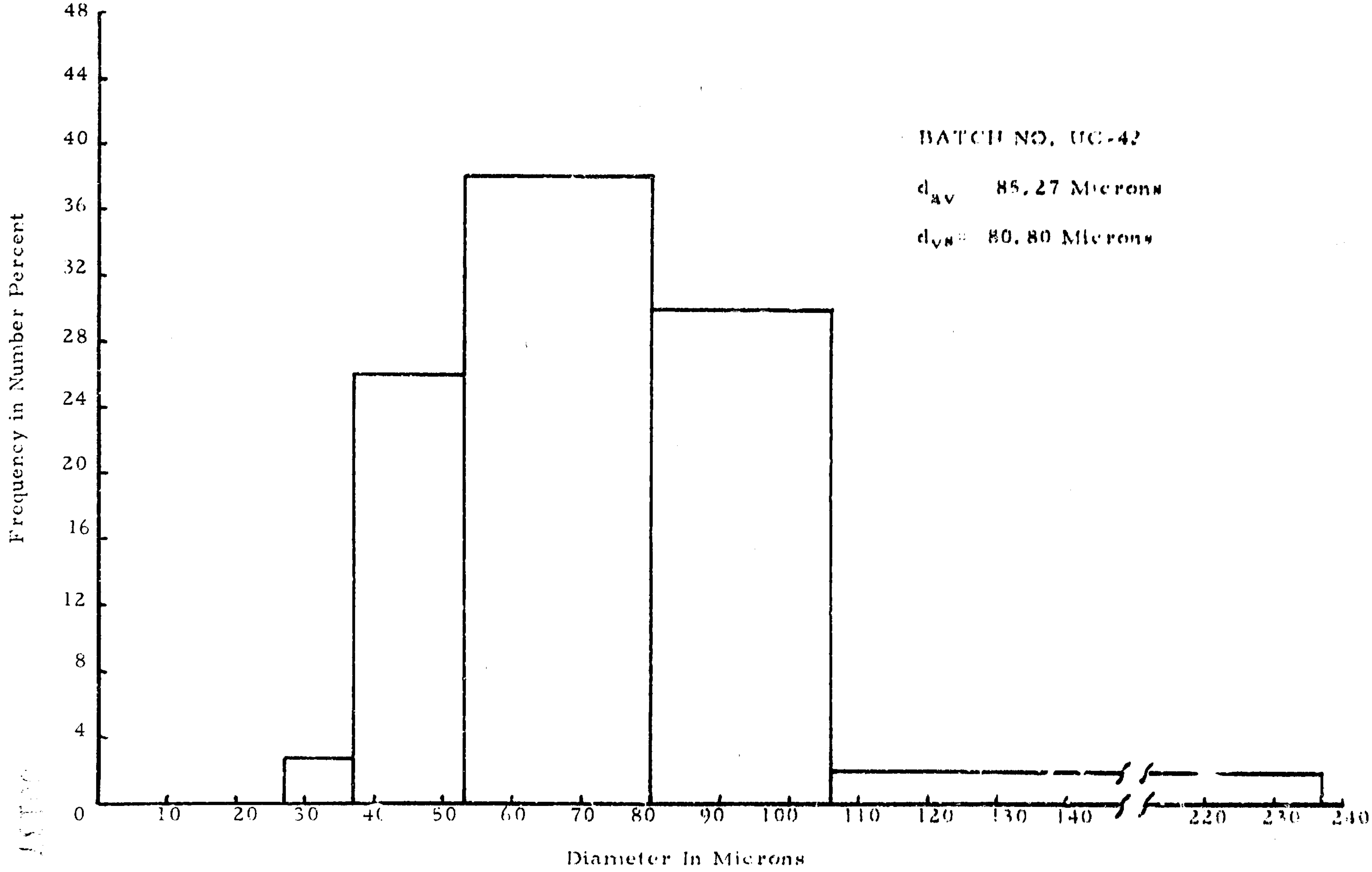

BATCH NO. UC-37
d_{av} = 77.38 Microns
d_{vs} = 55.37 Microns
FREQUENCY IN NUMBER PERCENT
0
4
8
12
16
20
24
28
32
36
40
44
48
10
20
30
40
50
60
70
80
90
100
110
120
130
140
150
160
170
180
DIAMETER IN MICRONS

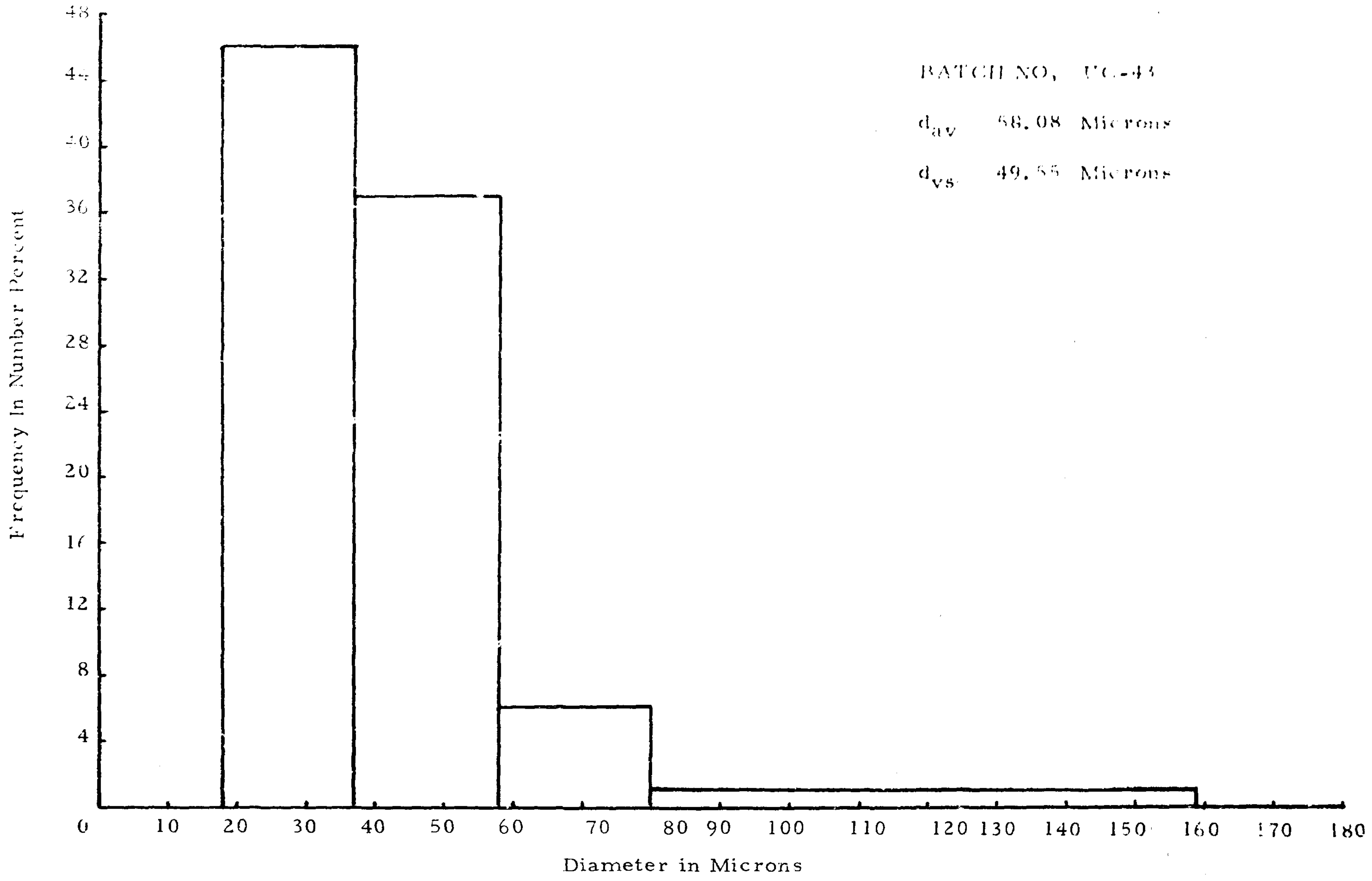

BATCH NO. UC-38
d_{av} = 60.99 Microns
d_{vs} = 51.22 Microns
48
44
40
36
32
28
24
20
16
12
8
4
0
10
20
30
40
50
60
70
80
90
100
110
120
130
140
150
160
170
180
DIAMETER IN MICRONS

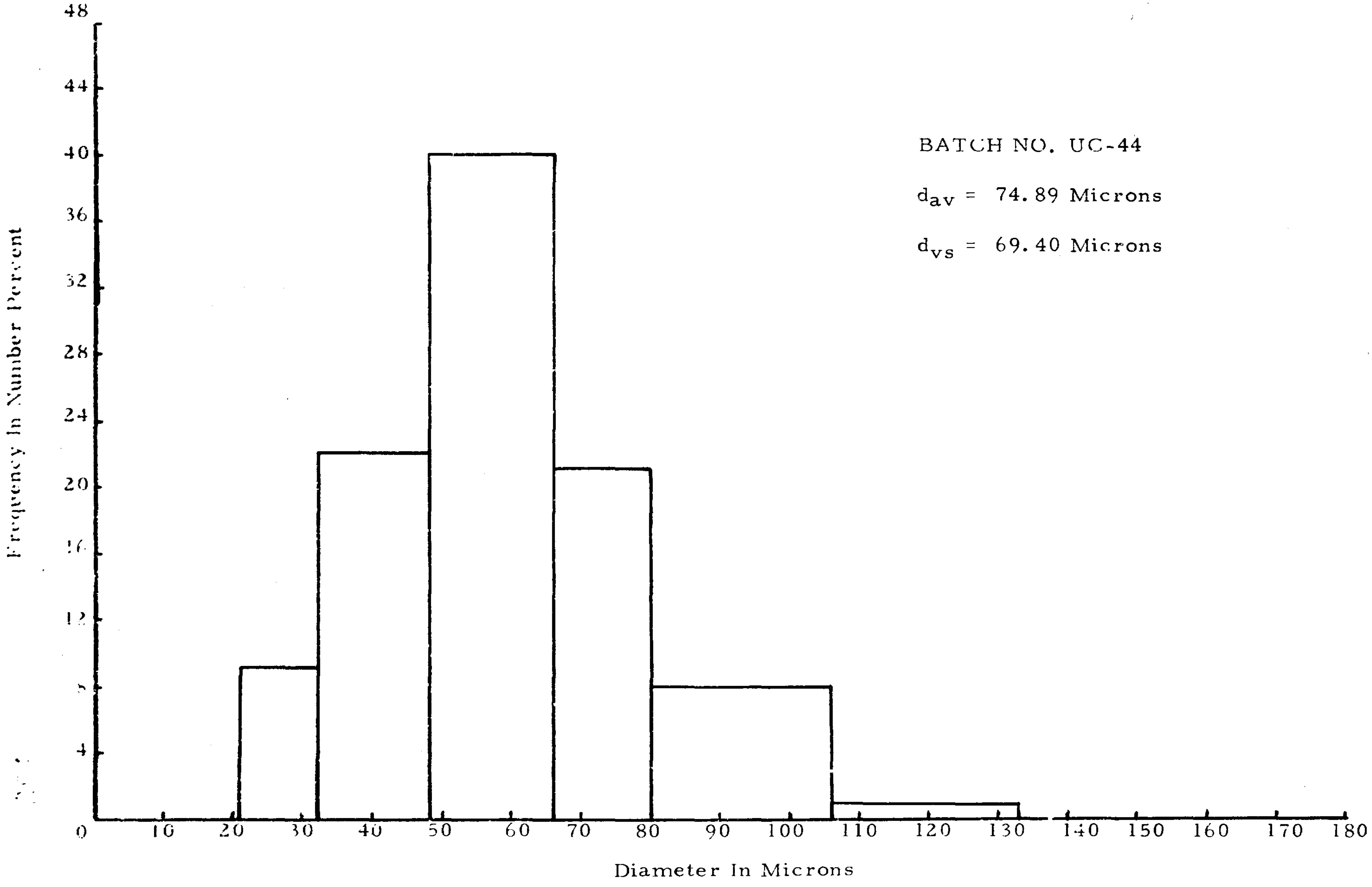

BATCH NO. UC-39
d_{av} = 85.22 Microns
d_{vs} = 82.82 Microns
Frequency in Number Percent
48
44
40
36
32
28
24
20
16
12
8
4
0
10
20
30
40
50
60
70
80
90
100
110
120
130
140
150
160
170
180
Diameter In Microns


BATCH NO. UC-40
d_{av} = 64.55 Microns
d_{vs} = 57.07 Microns
Frequency in Number Percent
48
44
40
36
32
28
24
20
16
12
8
4
0
10
20
30
40
50
60
70
80
90
100
110
120
130
210
220
230
240
DIAMETER IN MICRONS


BATCH NO. UC-41
d_{av} 94.46 Microns
d_{vs} 63.37 Microns
Frequency In Number Percent
48
44
40
36
32
28
24
20
16
12
8
4
0
10
20
30
40
50
60
70
80
90
100
110
120
130
140
220
230
240
Diameter In Microns

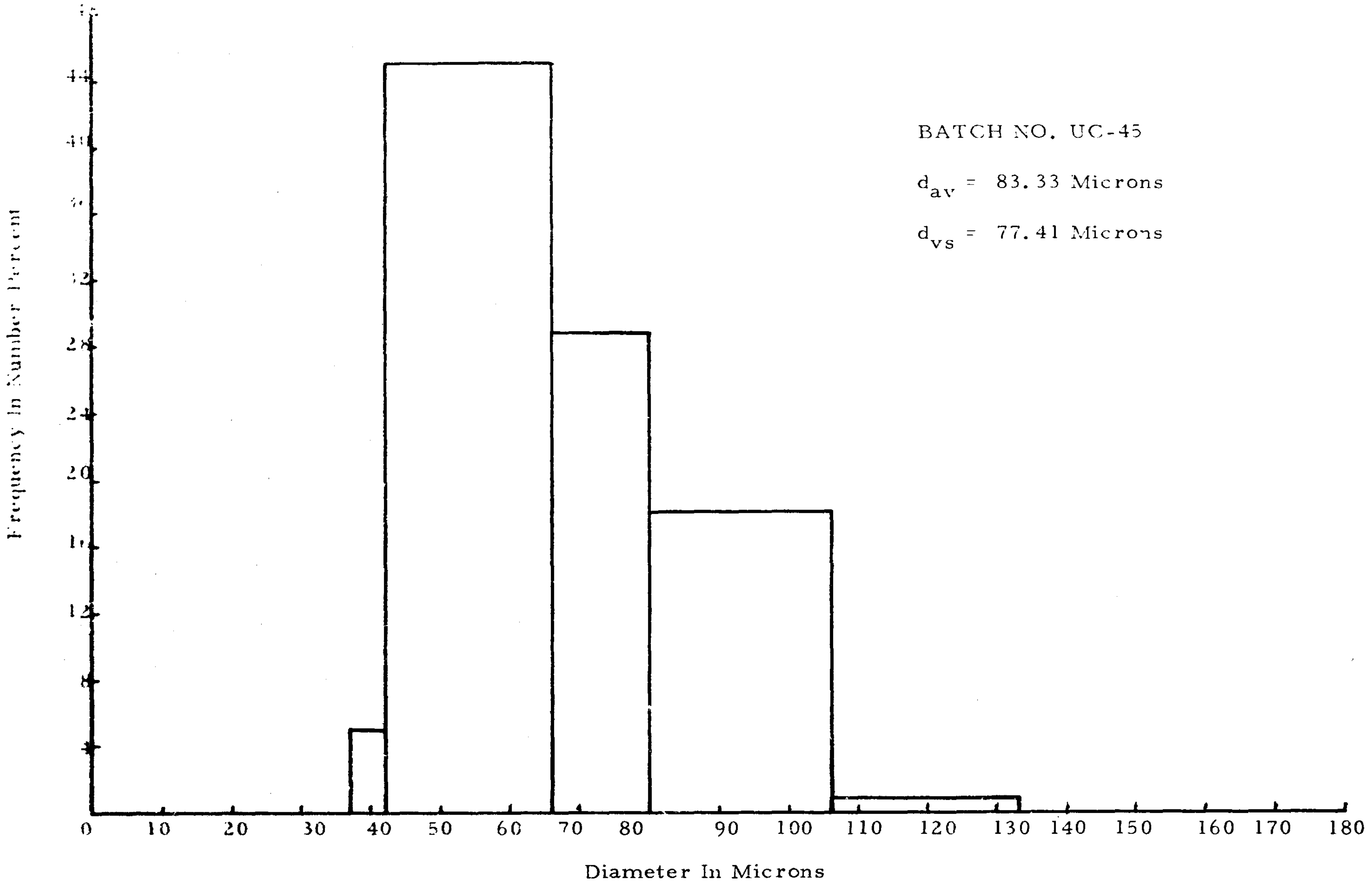

BATCH NO. UC-42
d_av 85.27 Microns
d_vn= 80.80 Microns
Frequency in Number Percent
0
4
8
12
16
20
24
28
32
36
40
44
48
10
20
30
40
50
60
70
80
90
100
110
120
130
140
220
230
240
Diameter In Microns

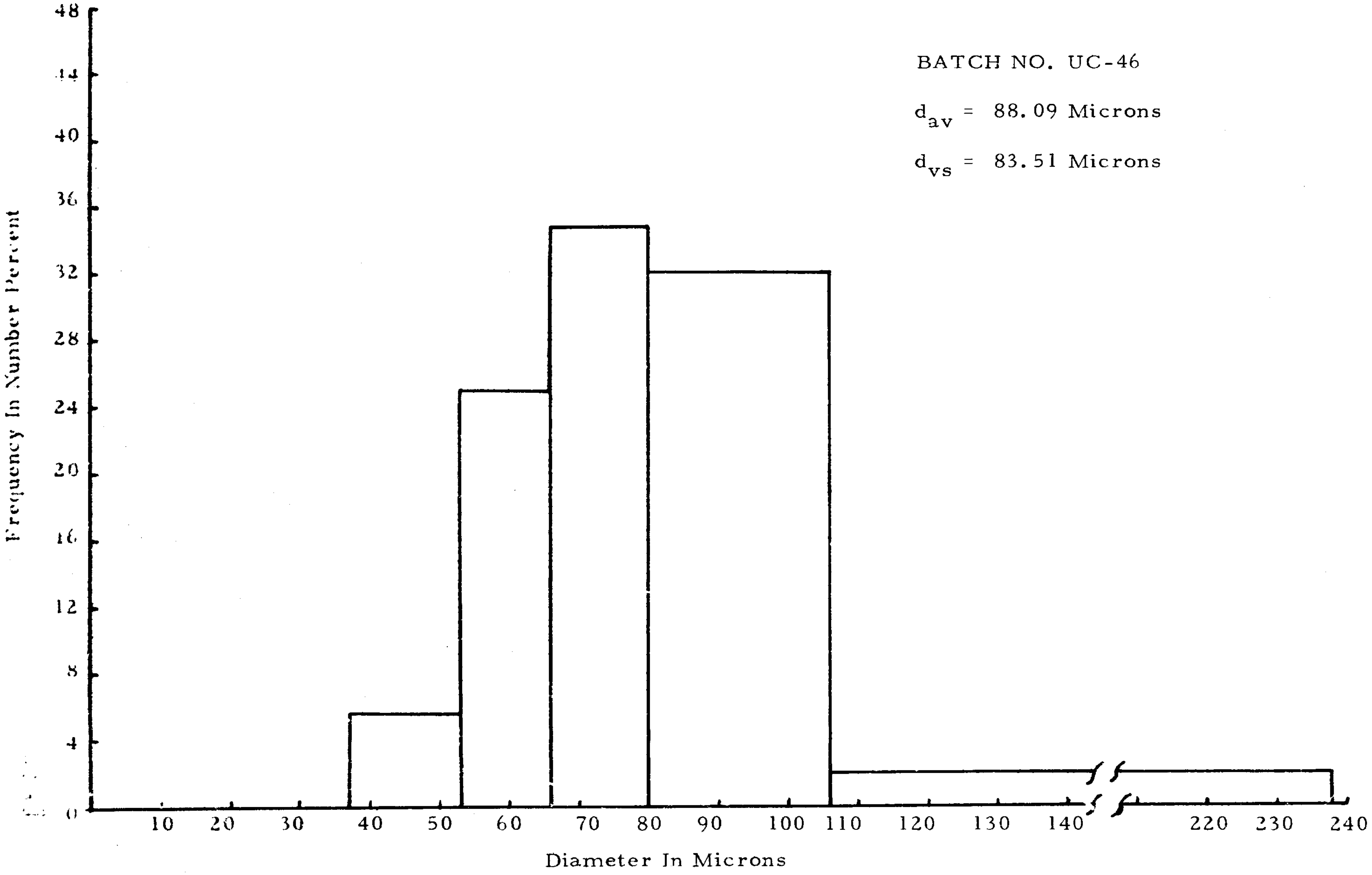

BATCH NO, UC-43
d av 58.08 Microns
d vs 49.55 Microns
Frequency In Number Percent
0
4
8
12
16
20
24
28
32
36
40
44
48
0
10
20
30
40
50
60
70
80
90
100
110
120
130
140
150
160
170
180
Diameter in Microns

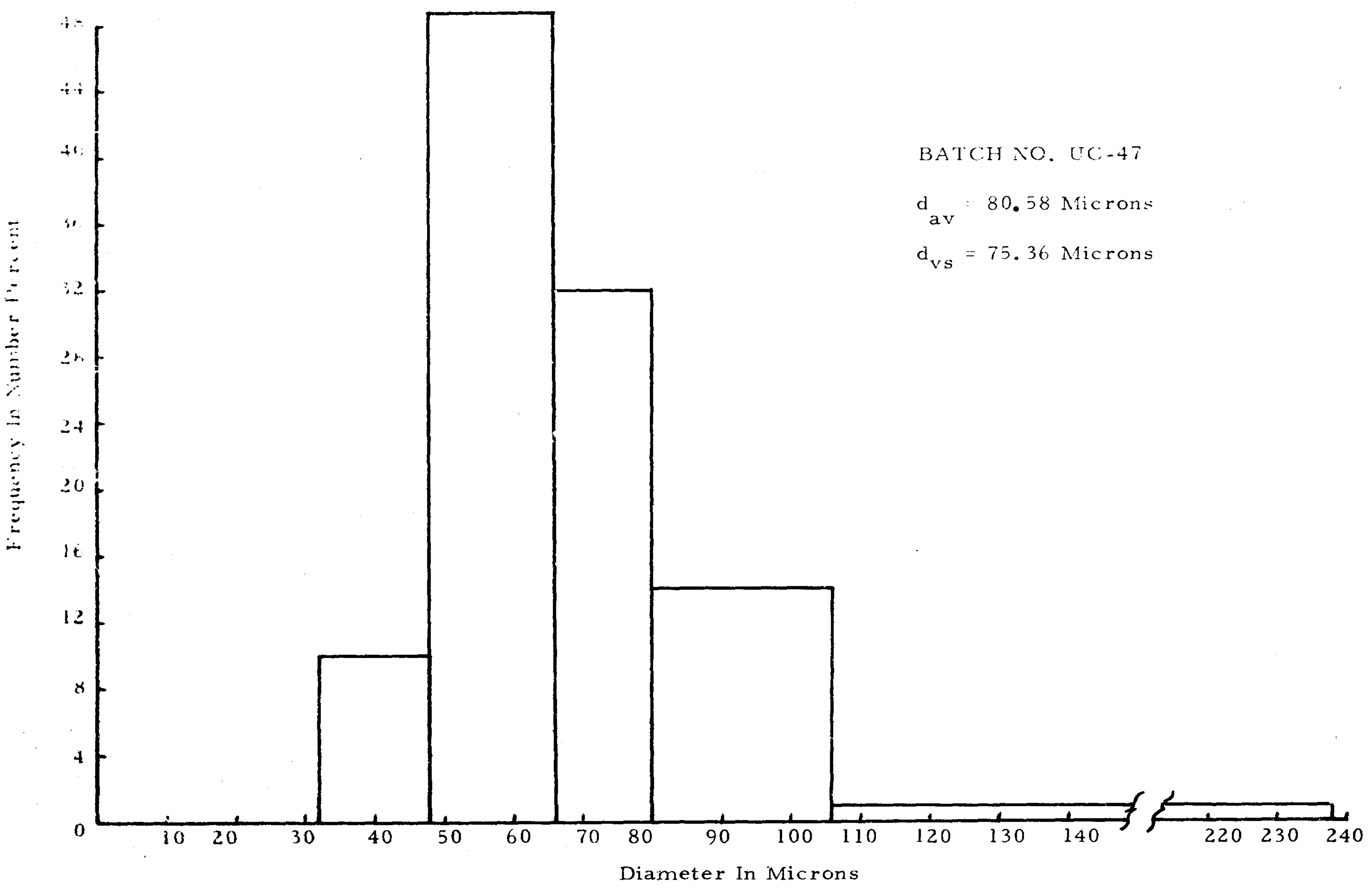

BATCH NO. UC-44

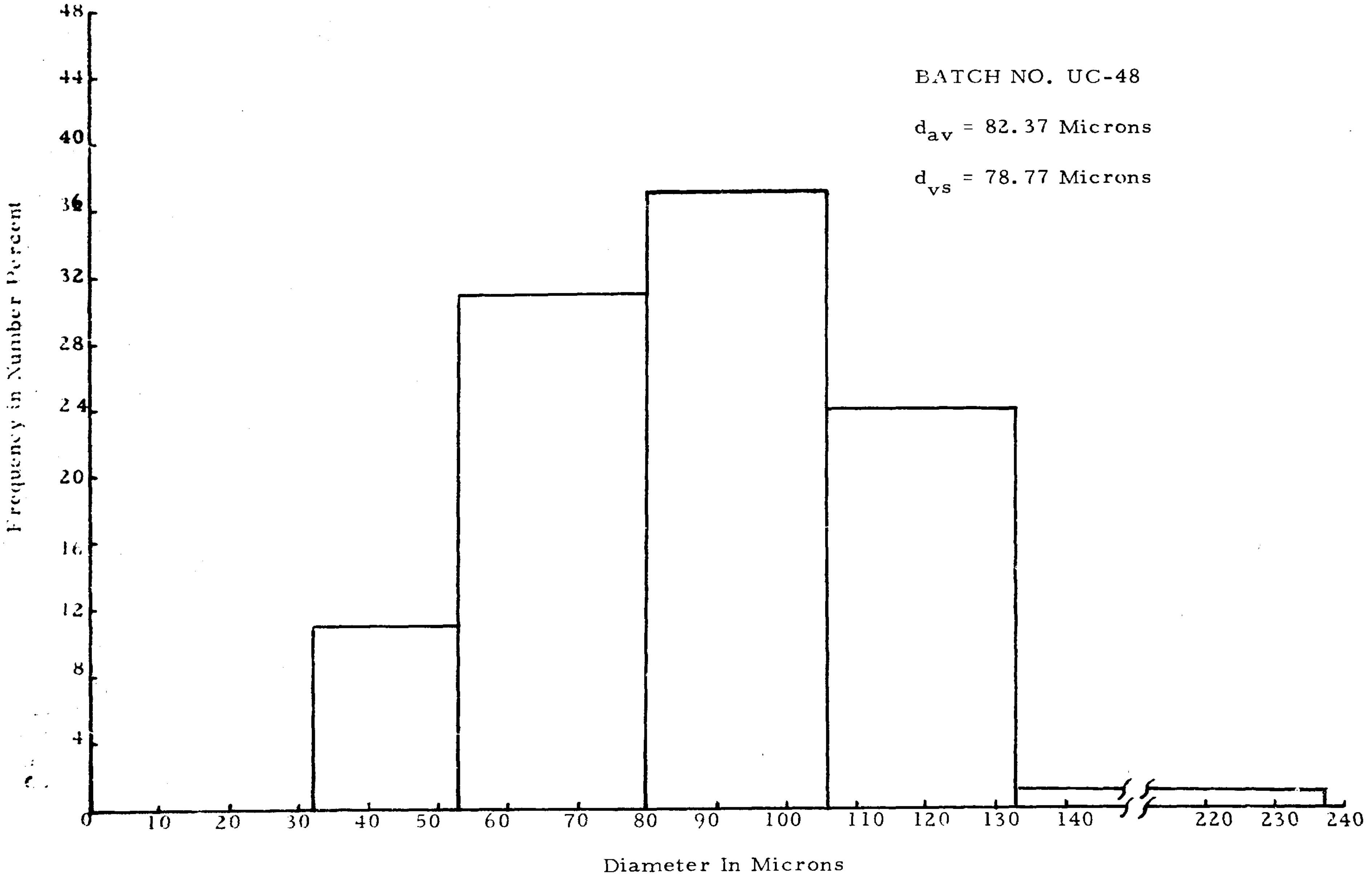
d_{av} = 74.89 Microns

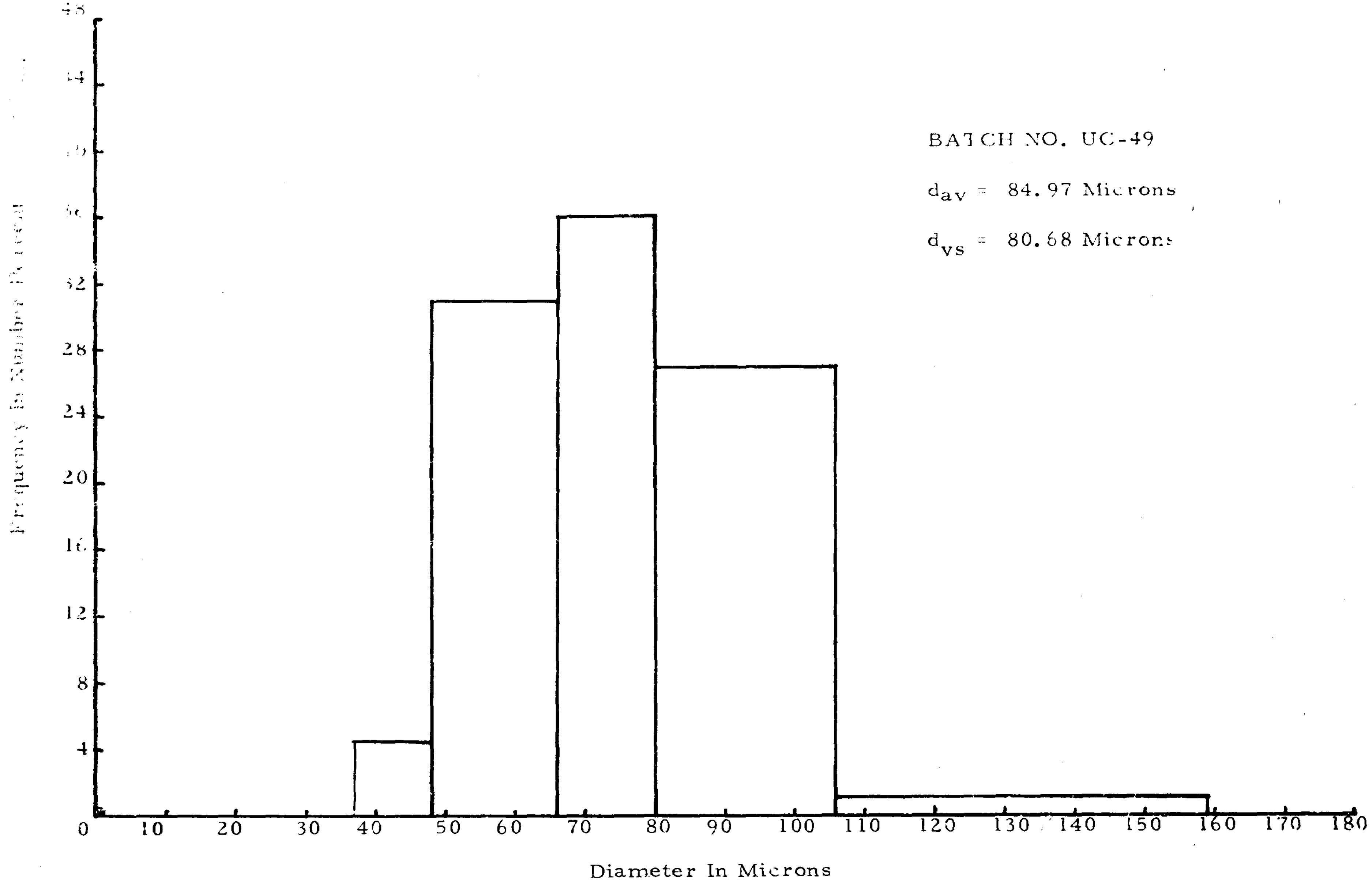
d_{vs} = 69.40 Microns

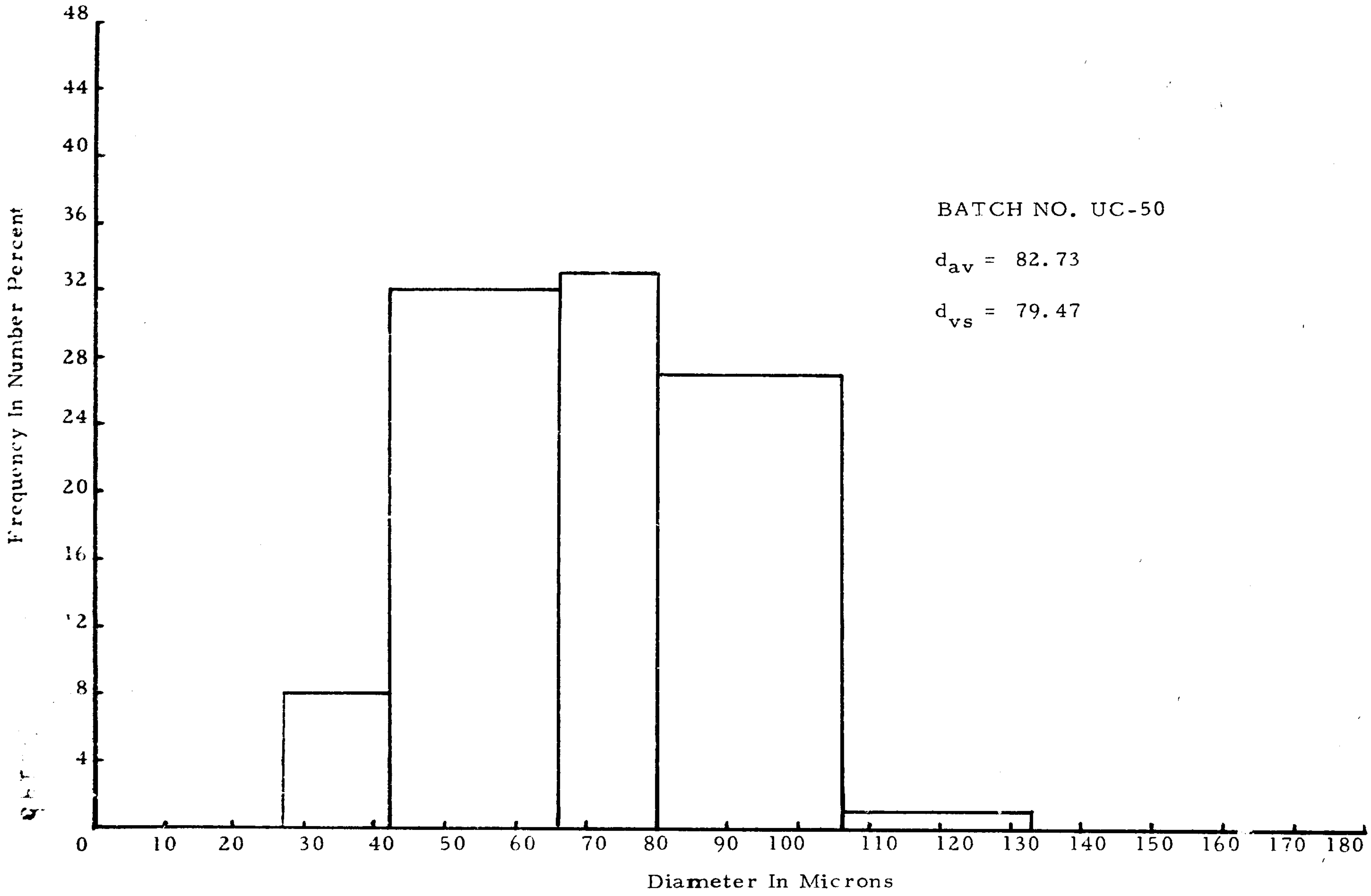

BATCH NO. UC-45
d_{av} = 83.33 Microns
d_{vs} = 77.41 Microns
Frequency In Number Percent
48
44
40
36
32
28
24
20
16
12
8
4
0
10
20
30
40
50
60
70
80
90
100
110
120
130
140
150
160
170
180
Diameter In Microns

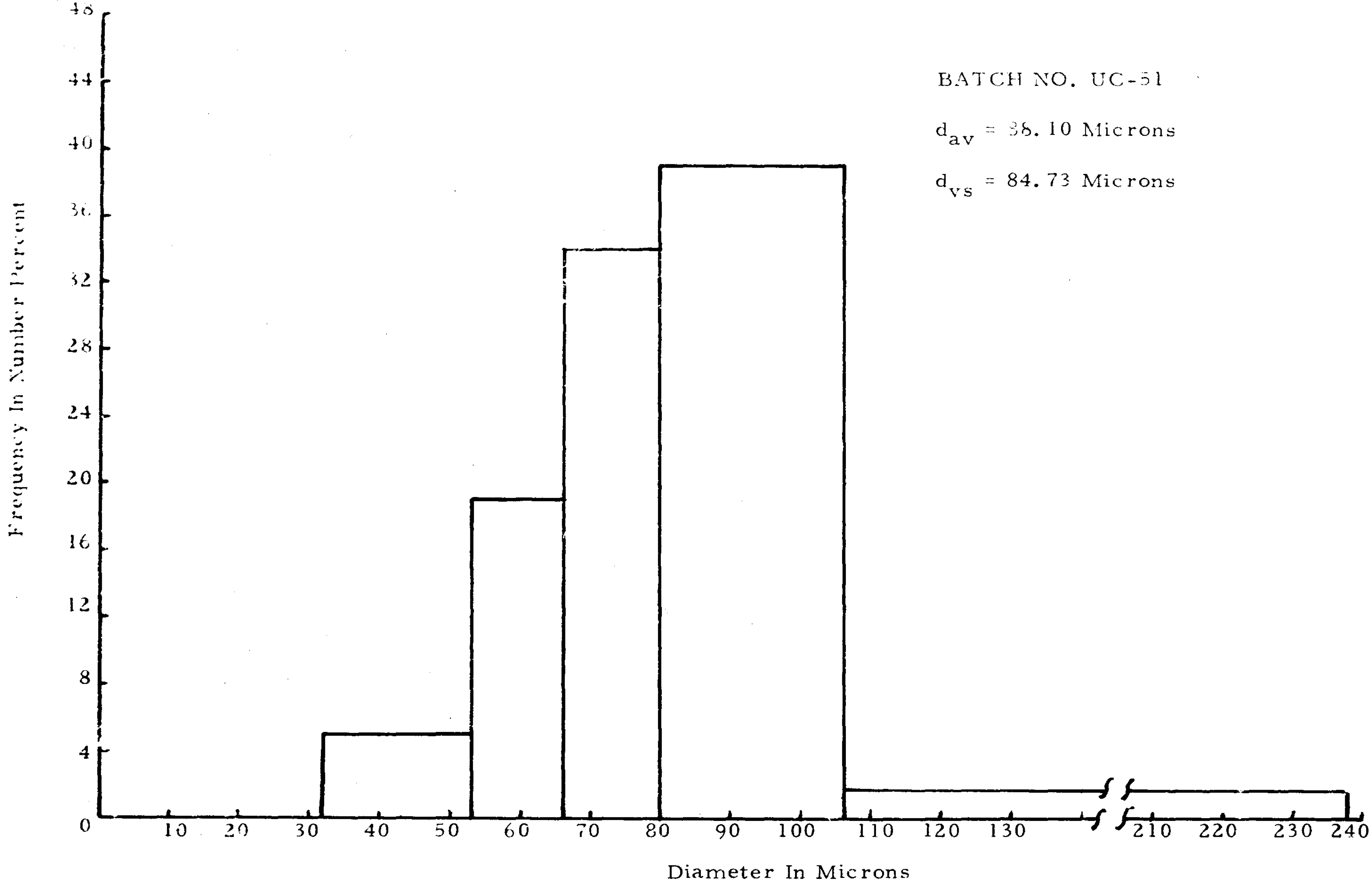

BATCH NO. UC-46
d_{av} = 88.09 Microns
d_{vs} = 83.51 Microns
Frequency In Number Percent
0
4
8
12
16
20
24
28
32
36
40
44
48
Diameter In Microns
10
20
30
40
50
60
70
80
90
100
110
120
130
140
220
230
240

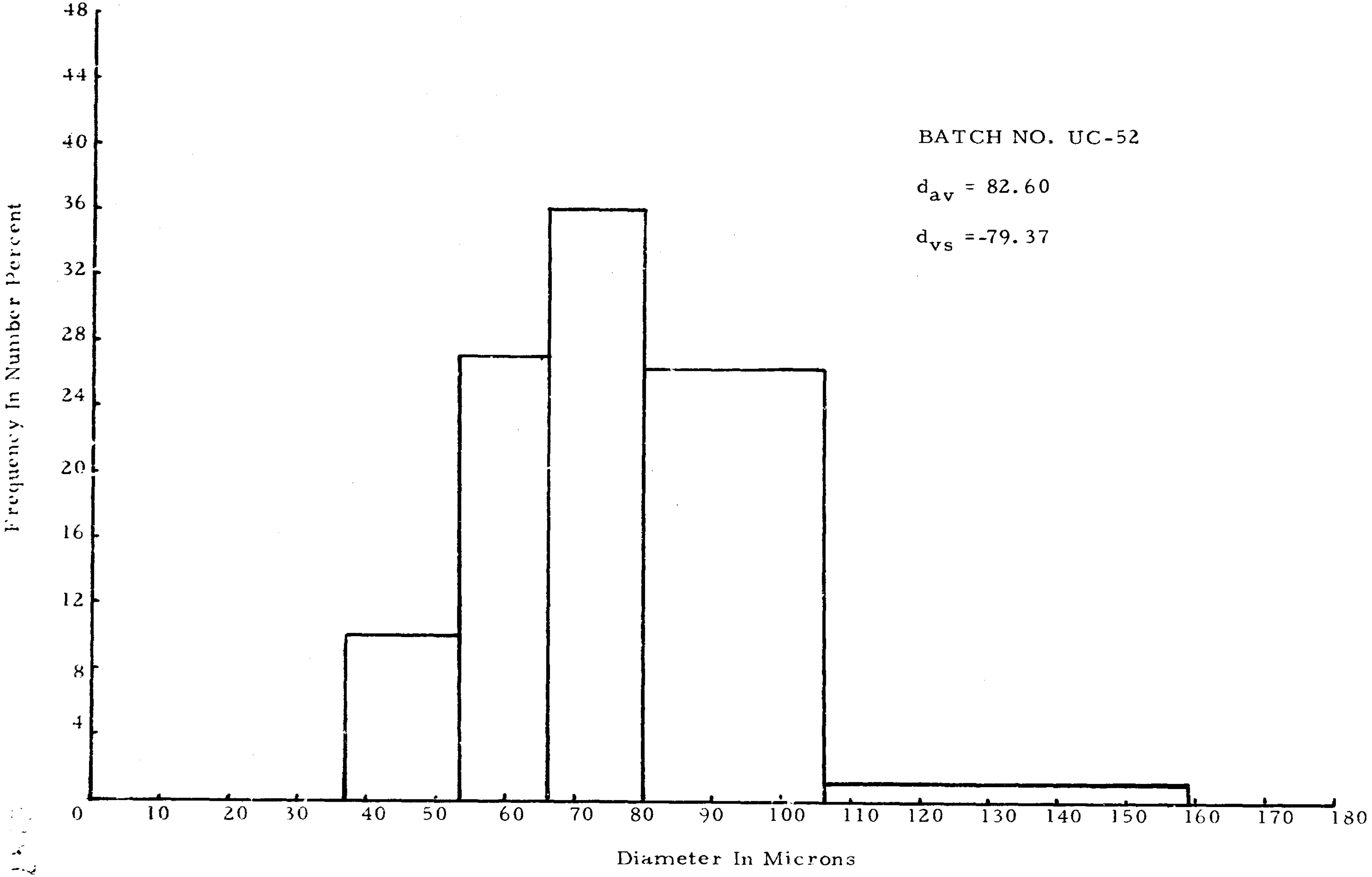

BATCH NO. UC-47
d_{av} = 80.58 Microns
d_{vs} = 75.36 Microns
Frequency In Number Percent
48
44
40
36
32
28
24
20
16
12
8
4
0
10
20
30
40
50
60
70
80
90
100
110
120
130
140
220
230
240
Diameter In Microns

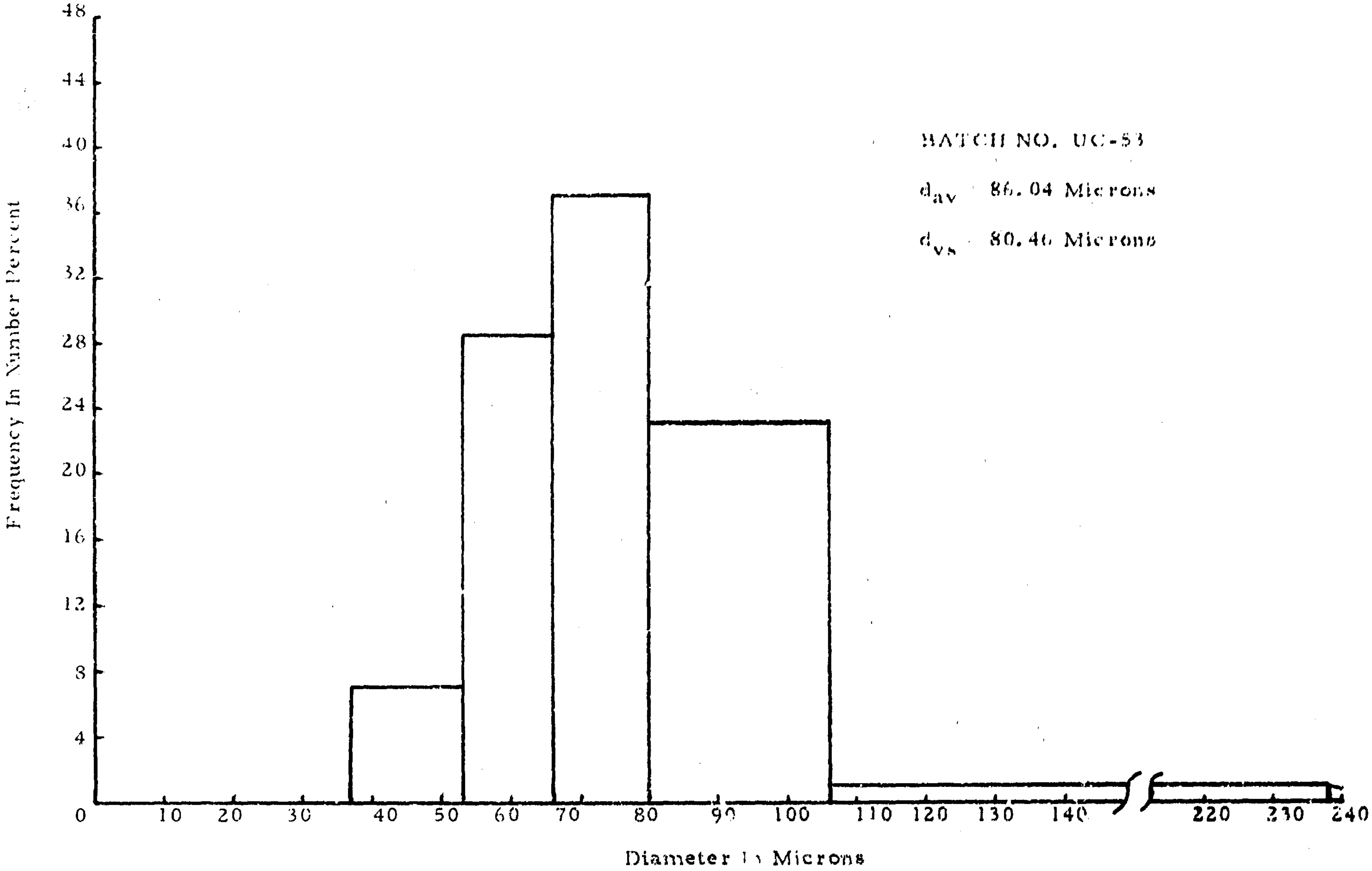

BATCH NO. UC-48
d_{av} = 82.37 Microns
d_{vs} = 78.77 Microns
Frequency in Number Percent
48
44
40
36
32
28
24
20
16
12
8
4
0
10
20
30
40
50
60
70
80
90
100
110
120
130
140
220
230
240
Diameter In Microns

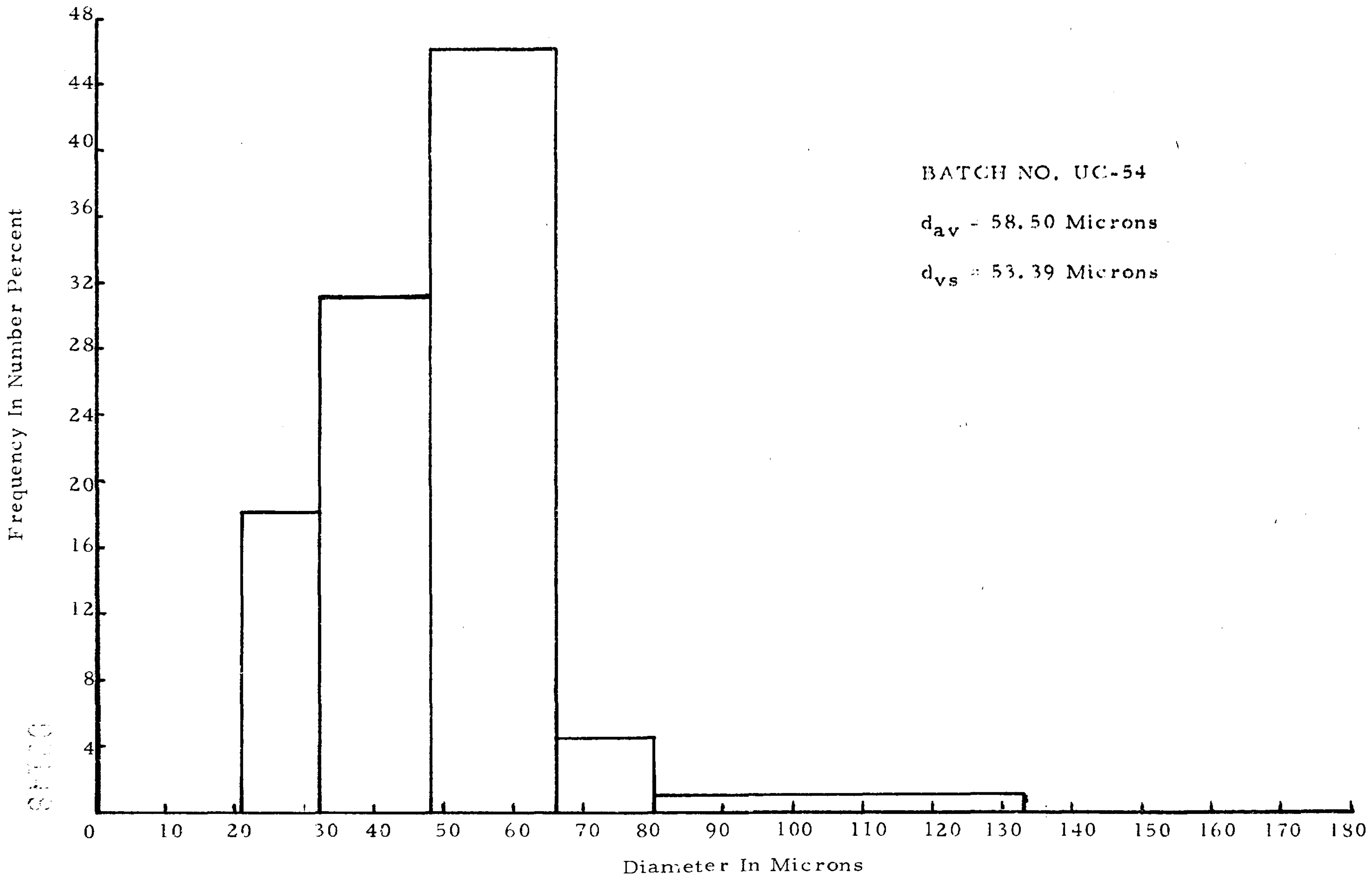

BATCH NO. UC-49
d_{av} = 84.97 Microns
d_{vs} = 80.68 Microns
Frequency In Number Percent
48
44
40
36
32
28
24
20
16
12
8
4
0
10
20
30
40
50
60
70
80
90
100
110
120
130
140
150
160
170
180
Diameter In Microns


BATCH NO. UC-50
d_{av} = 82.73
d_{vs} = 79.47
Frequency In Number Percent
48
44
40
36
32
28
24
20
16
12
8
4
0
10
20
30
40
50
60
70
80
90
100
110
120
130
140
150
160
170
180
Diameter In Microns


BATCH NO. UC-51
d_{av} = 38.10 Microns
d_{vs} = 84.73 Microns
Frequency In Number Percent
48
44
40
36
32
28
24
20
16
12
8
4
0
10
20
30
40
50
60
70
80
90
100
110
120
130
210
220
230
240
Diameter In Microns


BATCH NO. UC-52
d_{av} = 82.60
d_{vs} =-79.37
Frequency In Number Percent
48
44
40
36
32
28
24
20
16
12
8
4
0
10
20
30
40
50
60
70
80
90
100
110
120
130
140
150
160
170
180
Diameter In Microns

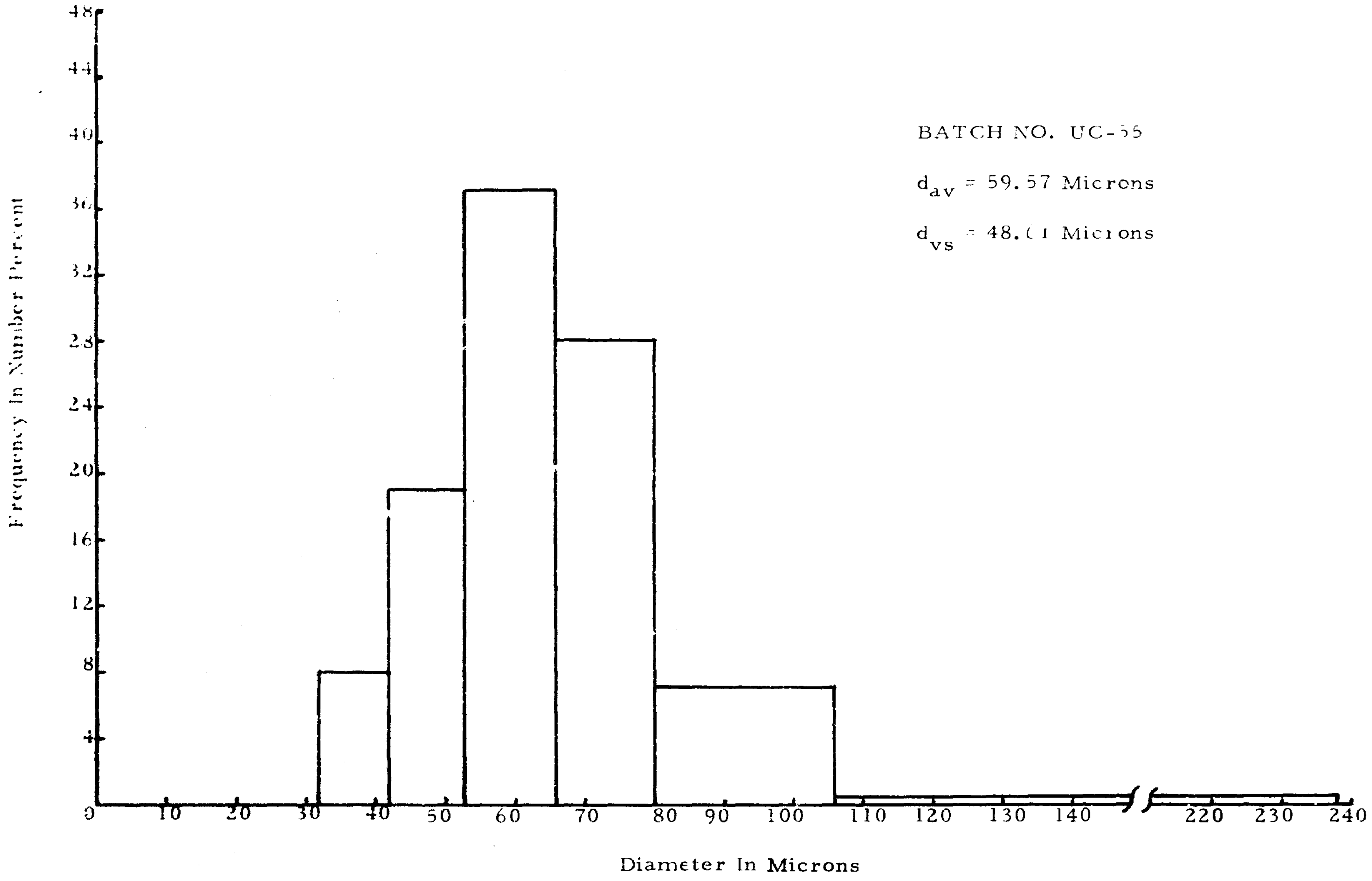

BATCH NO. UC-53
d_{av} 86.04 Microns
d_{vs} 80.46 Microns
Frequency In Number Percent
0
4
8
12
16
20
24
28
32
36
40
44
48
10
20
30
40
50
60
70
80
90
100
110
120
130
140
220
230
240
Diameter In Microns



BATCH NO. UC-54

d_{av} - 58.50 Microns

d_{vs} = 53.39 Microns

BATCH NO. UC-55
d_av = 59.57 Microns
d_vs = 48.01 Microns
Frequency In Number Percent
48
44
40
36
32
28
24
20
16
12
8
4
0
10
20
30
40
50
60
70
80
90
100
110
120
130
140
220
230
240
Diameter In Microns

APPENDIX III

SHIPPING PALLET COMPOSITIONS OF ENCAPSULATED UREA DELIVERED TO AFCRL

Pallet Compositions of Shipment Delivered September, 1970, to Meteorology Research, Inc., Altadena, California

Skid No.	Batch No.	No. of Cartons	Net Wt. (lbs.)	d_{av} of Batch (μ)*	d_{av} of Skid (μ)*
1	AP-432	12	300	66	66
	AP-434	13	325	65	
	UC-4	2	50	71	
2	UC-4	11	275	71	70
	UC-7	12	300	68	
	UC-8	4	100	70	
3	UC-8	9	225	70	67
	UC-10	14	350	66	
	UF-1	4	100	64	
4	UF-1	10	250	64	62
	UC-12	9	225	61	
	UC-13	8	200	61	
5	UC-13	4	100	61	58
	UC-14	13	325	59	
	UC-9	10	200	55	
6	UC-9	5	125	55	55
	UC-11	16	400	56	
	UF-5	6	150	53	
7	UF-5	8	200	53	50
	UF-6	13	325	49	
	UF-3	6	150	46	
8	UF-3	6	150	46	46
	UF-2	13	325	48	
	UF-4	8	200	42	
9	AP-433	13	325	90	94
	UC-6	13	325	98	
	UC-5	1	25	82	
10	UC-5	9	225	82	80
	UC-15	14	350	77	
	UC-17	4	100	86	

Total Net Wt. = 6,750 lbs.

*Arithmetic Mean Diameter in Microns

Pallet Compositions of Shipment Delivered November, 1970, to Meteorology Research, Inc. Altadena, California

Skid No.	Batch No.	No. of Cartons	Net Wt. (lbs.)	d_{av} of Batch (μ)*	d_{av} of Skid (μ)*
	UC-17	10	250	86	
11	UC-22	15	375	90	88
	UC-23	2	50	84	
	UC-23	13	325	84	
12	UC-28	13	325	80	82
	UC-18	1	25	77	
	UC-18	12	300	77	
13	UC-19	14	350	72	74
	UC-24	1	25	68	
	UC-24	13	325	68	
14	UC-26	13	325	64	66
	UC-20	1	25	62	
	UC-20	13	325	62	
15	UC-27	14	350	59	60
16	UC-19	2	50	72	62
	UC-29	13	325	62	
	UC-30	12	300	60	
	UC-30	2	50	60	
17	UC-38	9	225	60	61
	UC-25	14	350	60	
	UC-31	2	50	72	
	UC-38	5	125	60	
18	UC-31	13	325	72	71
	UC-37	9	225	77	
	UC-37	9	225	77	
19	UC-28	1	25	80	88
	UC-36	17	425	94	
	UC-36	7	175	94	
20	UC-21	14	350	96	95
	UC-34	6	150	95	

Total Net Wt. = 6,750 lbs.

*Arithmetic Mean Diameter in Microns

Pallet Compositions of Shipment Delivered December, 1970, to McClellan AFB, Sacramento, California

Skid No.	Batch No.	No. of Cartons	Net Wt. (lbs.)	d_{av} of Batch (μ)*	d_{av} of Skid (μ)*
21	UC-40	12	300	64	61
	UC-43	14	350	58	
	UC-44	1	25	75	
22	UC-44	19	475	75	77
	UC-47	8	200	81	
23	UC-47	8	200	81	82
	UC-48	15	375	82	
	UC-50	4	100	83	
24	UC-50	12	300	83	83
	UC-45	15	375	83	
25	UC-45	5	125	83	84
	UC-39	9	225	83	
	UC-42	13	325	85	
26	UC-42	3	75	85	86
	UC-49	16	400	85	
	UC-46	8	200	88	
27	UC-41	18	450	95	93
	UC-51	9	225	88	
28	UC-51	8	200	88	85
	UC-52	15	375	83	
	UC-45	2	50	83	
	UC-39	1	25	83	
	UC-46	1	25	88	
29	UC-53	17	425	86	83
	UC-46	1	25	83	
	UC-55	9	225	76	
30	UC-54	20	500	59	63
	UC-55	7	175	76	
31**	UC-35	18	450	117	120
	UC-33	2	50	144	
	UC-32	7	175	121	

Net Wt. of Skids 21-30 = 6,750 lbs.
Net Wt. of Skid 31 = 675 lbs.
7,425 lbs.

* Arithmetic Mean Diameter in Microns

** Skids 21-30, only, contain Tricalcium Phosphate additive.

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13. ABSTRACT

A process was developed for encapsulating hygroscopic, cloud-seeding agents, sodium chloride and urea. The encapsulation process involves depositing the coating polymer, ethylcellulose, onto finely divided powders using a phase separation-coacervation technique. The process produces small encapsulated aggregates, the size and distribution of which can be varied. Encapsulated materials prepared by the process are unique in that they are void, essentially, of powder fines. Laboratory tests of both encapsulated sodium chloride and urea proved the powders to be resistant to clumping and caking associated with premature moisture sorption occurring during storage and handling. The encapsulation coating polymer, ethylcellulose, is water insoluble, but is permeable to water vapor, water and solutes. Hence, water sorption and particle growth properties resulting from exposure of the encapsulated particles to humid atmospheres are effected by diffusion-mass transport processes. Laboratory testing of encapsulated sodium chloride and urea materials in simulated warm fog atmospheres revealed water sorption characteristics which were comparable to unencapsulated particles having equivalent dimension. Twenty thousand pounds of encapsulated urea, nominally, 20-80 microns in diameter, were produced and delivered for warm fog dissipation testing conducted by the Air Force Cambridge Research Laboratories. Results of the field tests show that the encapsulated urea functioned effectively and as designed.

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Warm Fog - hygroscopic nuclei						
Microencapsulation						
Sodium Chloride - microencapsulated						
Urea - microencapsulated						
Sizing Hygroscopic Powders - microencapsulation						
Encapsulation - ethylcellulose						
Coacervation						
Polymer Phase Separation						