MASTER

IS-334

Chemistry - General

SEP . 1961

**MASS TRANSFER COEFFICIENTS AND INTERFACIAL
AREA IN A ONE STAGE PULSE COLUMN**

By

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November, 1960

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Work performed under Contract No. W-7405-eng-82.

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MASS TRANSFER COEFFICIENTS AND INTERFACIAL AREA
IN A ONE STAGE PULSE COLUMN*

By

Alvin Eugene Konopik and Lawrence Burkhardt

ABSTRACT

Over-all mass transfer coefficients were determined by considering the operation of the pulse column to be a stage-wise process. The area for mass transfer was obtained by direct photographic measurements. During the downstroke the aqueous phase passed through the organic phase in the form of rivulets as the aqueous phase preferentially wet the plate material. Because of these rivulets, the area for transfer during the downstroke could not be measured. Thus the calculated coefficients describe only the operation of the pulse column for the free rise of organic droplets through a stage. Variation in experimentally determined concentrations necessitated the calculation of three concentrations by material balance equations. The driving force for transfer was based on the aqueous phase concentrations.

The mass transfer coefficient was found to increase when the interfacial tension was reduced. The results of this study and those of Coulson and Skinner (5) are in agreement

*This report is based on an M. S. thesis by Alvin Eugene Konopik submitted November, 1960, to Iowa State University, Ames, Iowa. This work was done under contract with the Atomic Energy Commission.

on the effect of interfacial tension.

Two factors appear to affect the mass transfer coefficient as the pulse frequency is changed. These factors are: a) recycle or backmixing which tends to decrease the coefficient through a reduction of acetic acid in both phases and thereby increases the interfacial tension, and b) turbulence which enhances the coefficient by reducing the continuous film resistance. The first is important at or near the lower flooding limit, but the latter is the controlling factor at higher frequencies.

The H.T.U. concept is a useful tool in determining the over-all extraction efficiency. However, the method of analysis is not applicable for obtaining individual effects on the mass transfer coefficient and the mass transfer area. More fundamental principles probably could have been attained by Chu, et al. (3) in their work with wetting agents, which was discussed previously, if the interfacial area had been measured.

INTRODUCTION

The field of liquid-liquid extraction has grown to one of considerable importance in recent years because of the necessary separations required in the chemical industry. Although many types of extractors are in general use today, the mechanically agitated extractors are becoming more prominent because of their high efficiencies.

One of these extremely efficient extractors is the pulse column. It was first introduced by Van Dijck (19) in 1935. The original column was composed of pulsed plates but he noted that the same effect could be produced by mechanically pulsating the fluid. The latter method seems to be more prevalent because the maintenance costs are generally lower. A schematic representation of this mode of operation is shown in Figure 1. A unique feature of this column is that both of the light and heavy phases are alternately changed from the dispersed to the continuous phase as they are forced upward or downward through the column.

General interest in the pulse column occurred when it was first introduced for production at Hanford, Washington. Sege and Woodfield (15) noted that the two main advantages of the pulse column over the conventional packed column were that the extractor height was greatly reduced and temporary shut downs were made with greater ease. They divided the pulse column operation into five general regions as illustrated in Figure 2:

Figure 1. Diagram of a pulse column

Figure 2. Operating characteristics of a pulse column

- A) a region of complete flooding because of insufficient pulsation,
- B) a region of mixer-settler type operation, characterized by stable operation and separation of phases between strokes,
- C) a region of emulsion type operation, characterized by a high holdup of the dispersed phase and usually good extraction due to intimate contact of both phases,
- D) a region of unstable operation because of local flooding, and
- E) a region of complete flooding due to emulsification.

The only useful regions of operation are regions B and C.

Edwards and Beyer (6) studied pulse column flow rates with respect to flooding and concluded that flooding due to excessive pulsation was very difficult to describe. A mathematical expression was developed to predict the lower flooding point. Holdup of the discontinuous phase and general column performance were reported to be very sensitive to plate-wetting characteristics.

Treybal and Dumoulin (18) found that the design of the plate had a very profound effect on the allowable solvent throughput. The H.T.U. (height of a transfer unit) was observed to decrease sharply with decreased plate spacing to a point, and then was relatively constant for even smaller plate spacing. Griffith, et al. (8) state that smaller plate

spacing enhances the efficiency but reduces the pulse column throughput. The effects of plate spacing, hole diameter, percent free plate area, pulse amplitude, and pulse frequency on the extraction efficiency were investigated by Burkhart and Fahien (1). The recycle which exists in a pulse column was postulated to have an undesirable effect upon the extraction efficiency. Li and Newton (12) correlated their data for the toluene-benzoic acid-water system using the H.T.U. concept. The consensus of these investigators (1, 8, 12, 18) was that the H.T.U. increased with increasing frequency.

General equations and nomographs for flooding velocities and over-all H.T.U. based on the continuous phase for several systems were presented by Smoot, et al. (16). The two equations are in the form of dimensionless groups and the constants were determined by multiple regression techniques. These equations apply to operation in the emulsion region and to systems where the major resistance to transfer is in the dispersed phase film. Thornton (17) offered a design procedure based upon a limited number of laboratory runs. His work showed that a column could be designed to ± 15 percent in the range in which the tests were made by using the laboratory data and equations developed for calculating the column diameter and over-all H.T.U. based on the continuous phase.

Later, Burkhart and Fahien (2) presented a design procedure in which a stagewise approach was utilized rather

than the H.T.U. concept. A stage was defined as the portion of the column between two adjacent plates. A sampling procedure was outlined for obtaining internal column samples. These researchers proved experimentally that the concentration at some fixed point in the column depended upon the time of pulse cycle. The theory proposed that the number of stages could be stepped off between "operating" and "stage" lines much the same as on a McCabe-Thiele diagram. The theory and experimental data showed that it was possible to have two sets of these "stage" and "operating" lines. The experimental data were reported as upstroke and downstroke efficiencies. These efficiencies were analogous to Murphree plate efficiencies.

In his study of the effects of pulse column variables and system physical properties on droplet size, Graham (7) stated that the droplet size became larger with increased frequency at the lower flooding point but became smaller with increasing frequency under recycle conditions. Greater variation in droplet size was discovered at the lower frequencies than at the higher frequencies. A method of setting the physical size of the pulse column was proposed.

Coulson and Skinner (5) investigated the mechanism by which mass transfer takes place from a continuous phase to single droplets. These data were obtained for three conditions: (a) during droplet formation, (b) during droplet rise

through a stationary phase, and (c) during droplet rise through a downward moving phase. The over-all mass transfer coefficient (based on the average interfacial area of formation) decreased with increasing time of formation, but was practically independent of the droplet size. However, the mass transfer coefficient during rise through a continuous phase was found to increase as the droplet size became larger. These results substantiate those of Licht and Conway (13) who also studied the mechanism of mass transfer during droplet formation and during the fall of droplets through a continuous phase in a spray column.

SCOPE OF INVESTIGATION

Mass transfer coefficients in most of the existing literature are actually a product of a mass transfer coefficient and the contact area per cubic foot of extractor equipment. This product is not a logical grouping when considering the factors which affect extractor efficiency. There is no reason to believe that the physical properties of a system and the operating characteristics of an extractor affect the mass transfer coefficient and the interfacial area in the same manner. Rather the grouping of the terms was a convenience, as the area of interfacial contact between the two phases usually is not known.

The H.T.U. concept cannot be used to reveal separate effects on the mass transfer coefficient and the interfacial area because the product of the mass transfer coefficient and the interfacial area is used in its formulation. The investigators (1, 8, 12, 18) reported increases in H.T.U. with increasing pulse frequency. This indication of a greater resistance to mass transfer with improved agitation probably is caused by a combination of effects which are difficult to separate.

The present study was undertaken to separate the mass transfer coefficient from the interfacial area. In previous investigations by Licht and Conway (13) and Coulson and Skinner (5) droplet size was measured by the droplet volume

method. To determine the interfacial area by this method, one must assume that the droplets are spherical, know the number of droplets, and measure the volume of all the droplets. The droplet volume method could not be used in the present work because the number of droplets cannot be counted in a pilot plant extractor.

The droplet size was determined by the photographic technique outlined by Graham (7). This investigation was, perhaps, the first utilization of the photographic technique to obtain over-all mass transfer coefficients for the free rise of dispersed droplets in a pulse column.

The internal flow rates were calculated using the equation developed by Edwards and Beyer (6). Likewise, the sampling technique and stagewise approach presented by Burkhardt and Fahien (2) were used.

Briefly, the data for this investigation were taken with a 3 inch, glass pulse column in which the systems methyl isobutyl ketone-acetic acid-water and isophorone-acetic acid-water were used. The pulse amplitude was set at 1.252 cm. for the methyl isobutyl ketone-acetic acid-water system and 2.553 cm. for the isophorone-acetic acid-water system. The plates were spaced 3 inches apart for all runs. Only the effects of frequency, aqueous flow rate, and concentration on the mass transfer coefficient were considered.

Definitions of Run, Series, and Group

The distinction between run, series, and group was made to eliminate cumbersome repetition and to contribute to the continuity of the discussion. These definitions will be used throughout the remainder of this paper.

Run

Run is the distinction given to an individual measurement for concentration or holdup taken at a specific time during the pulse cycle.

Series

A complete set of runs in which the necessary data are obtained to calculate one mass transfer coefficient (designated by a group letter followed by a series number; i.e., B-2).

Group

A number of series where only one condition was varied; i.e., frequency, aqueous feed rate, concentration and number of stages (designated by a letter).

The following is an incultation of the distinction between run, series, and group.

The data necessary for the calculation of a mass transfer coefficient could not be obtained in one run. Because three

column isolations were required, a set of runs performed at identical conditions were referred to as a series. A number of these series in which only one condition was varied (e.g., frequency) were referred to as a group. Therefore, in groups A and B only the frequency was varied. A-1 and B-1 were both at the same pulse frequency of 15 cycles per minute. Series A-1 was obtained in a one stage column, while series B-1 was obtained in a two stage column.

Groups A, B, F, G, and H were performed to determine the effect of pulse frequency on the mass transfer coefficient. The purposes of groups C, D, and E were to determine the effect of aqueous flow rate, aqueous feed concentration, and the position of the stage in the column, respectively, on the mass transfer coefficient.

An explanatory arrangement of the foregoing definitions is shown below:

Run	Series	Group
end of: downstroke (concentration) upstroke (concentration) downstroke (holdup)	A complete set of data to calculate one mass transfer coefficient	A number of series in which only one condition was varied; i.e., frequency, aqueous feed rate, concen- tration
_____	_____	
_____	_____	
_____	_____	
_____	_____	

EXPERIMENTAL MEASUREMENTS

Equipment and Pulse Characteristics

A schematic diagram of the pulse column and related equipment is presented in Figure 3. A photograph of a two stage pulse column is shown in Figure 4. The pulse column was made of pyrex glass sections, 3 inches in diameter and 3 inches high. The plate material used was 316 stainless steel which was perforated to the specifications of 0.033-inch hole diameter and a 21 percent free area by the Harrington and King Perforating Company. These plates were sandwiched between two polyethylene gaskets and the glass sections. The whole assembly was compressed until a seal was maintained with four tie rods which extended the length of the column.

Aqueous feed was fed to the top column by an Eastern model D-11 centrifugal pump and metered through a Brooks rotameter equipped with a Teflon float. Organic feed was fed to the bottom of the column by a Milton Roy Constametric pump regulated by a General Electric Tymotrol speed control unit. This flow rate was governed by a tenturn Helipot which was calibrated to give the desired flow rate. The raffinate stream was pumped out of the column by an Eastern model D-11 pump and metered through a Brooks rotameter equipped with a tantalum float.

The extract stream was taken from the top of the column through an overflow line and measured in a graduated cylinder

Figure 3. Flow diagram of the pulse column and auxiliary equipment

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for 2 to 3 minutes. The raffinate stream was measured in the same manner.

The pulse amplitude was measured with a pointed stainless steel probe attached to a micrometer vernier which was mounted on the top of the column. The difference in readings, taken when the point of the probe just touched the interface at the top of an upstroke and when the interface just broke away from the probe at the bottom of the downstroke, was added to the correction for wetting of the probe by the aqueous phase at the bottom of the downstroke. The correction was determined by stopping the pulse generator, lowering the probe until it just touched the interface, and then raising it until the interface just broke away from the probe. The value of the pulse amplitude by the method utilized was checked using a short-range telescope.

The pulse generator consisted of a 6 inch diameter stainless steel bellows driven by a variable speed drive through a gear box from the Lapp Pulsafeeder. The pulse frequency was timed three times for one minute during each individual run. The stripping column consisted of two sections of 4 inch diameter, heavy duty pyrex glass, each 5 feet long, flanged together and packed with 0.25 inch Berl saddles. The extract from the pulse column was fed countercurrently to distilled water in the stripping column. The acid content in the organic product was reduced below the necessary concentration

for solvent reuse in the pulse column. Acid was added to this product until the desired acid concentration in the organic feed was attained. In addition to the acid already present in the aqueous product, more acid was added to obtain the desired aqueous feed concentration to the pulse column.

Measurement of Holdup

The organic phase holdup was measured with a short range traveling telescope after the column was isolated by the valving mechanism shown schematically in Figure 5. The mechanism was used to isolate the aqueous feed, raffinate, and pulse streams at any desired time during the pulse cycle. Since the Milton Roy Constametric pump can develop an outlet pressure of 6000 psi., a valve could not be put in the organic feed line. However, the switch that turned off the current to the pump also closed an electrical circuit which made the pump act as an electrical generator. Thus the momentum of the fluid was stopped by the pump as well as if a valve had been placed in the organic feed line. The individual stage holdup could be measured because the organic phase did not pass through the plate due to interfacial tension.

Measurement of Concentration

Each glass section of the pulse column was equipped with a sample port sealed with a rubber ampule plug. Internal

300

Figure 5. Valving mechanism for column isolation

column concentration samples were taken through "L" shaped stainless steel hypodermic needles which had machined tips that fit snugly through the plate perforations. Figure 6 is an illustration showing the construction which is described above.

With the needle in the upward position, an aqueous sample could be removed from the plate above during the downstroke of the pulse cycle. The concentration of this sample corresponded to the aqueous concentration at the end of an upstroke. By reversing the conditions described above, an organic concentration at the end of a downstroke was obtained from the plate below the rubber ampule plug. Internal samples were taken only after the end product material balances checked within ± 3.0 percent. The samples were removed through 0.125 inch rubber tubing connected to sample bottles through solenoid control valves. The procedure further required that two or three droplets of sample be taken during each cycle until enough was collected to be analyzed.

Since the organic phase at the end of an upstroke and the aqueous phase at the end of a downstroke do not go through the plates during column operation, samples were removed after column isolation similar to that required for holdup measurements. The above samples, however, were taken through 1/8 inch needles, which permitted the removal of the necessary sample size in one second or less after column isolation.

Figure 6. Detail of the sampling technique

When the pulse column was not operated near the lower flooding point, the organic phase at the end of a downstroke could not be obtained during column operation since two phases came through the plate. It then became necessary to obtain this sample after column isolation. All samples were titrated with a Beckman titrimeter.

Measurement of Droplet Size

The droplet diameters were determined by photographing the droplets, mounting the negatives as slides, projecting these slides on an 8 x 8 inch ground glass screen of a Kodaslide table viewer, and measuring the droplets directly from this screen with a transparent ruler. Photographs were taken with a Leica 35 mm. model III-g camera with a G extension tube and an f-3.5 lens. The light source was a Heiland strobe flash unit located directly behind the column facing the camera. The arrangement of the photographic equipment is shown in Figure 7. The lens opening was set at f-22 and the camera shutter speed was set at 1/30 second. Since the laboratory light was negligible compared to the strobe light source, the effective shutter speed was 1/2000 of a second. A plastic spacer guide kept the distance from the camera to the column the same for every picture. The constant magnification factor of 3.15 was insured by using this guide.

The diameter of a sphere having the average specific

Figure 7. Arrangement of the photographic equipment

surface, d_{vg} , was calculated from 25 droplets selected randomly from each picture. The method of calculation was outlined by Graham (7). Values of d_{vg} reported are average d_{vg} 's determined from three or more photographs.

RESULTS

The experimental data are shown in Appendix C. A complete series of data for a unique set of operating conditions required at least three separate experimental runs. (Runs, series, and groups are defined on page 10). Therefore, the reported data are average values for some measurements and smoothed points for others.

The values of pulse frequency, feed and product flow rates, feed and product concentrations, and droplet size are averages of the individual run measurements (11) for all groups A through H. Typical variations from these averages are shown in Tables 1 and 2.

Table 1. Variation of pulse frequency and feed and product flow rates for series B-2

Run No.	Pulse Frequency cpm.	Flow rates, ml./min.			
		Aqueous feed	Raffinate	Organic feed	Extract
1	19.76	545	455	750	821
12	20.06	545	444	750	845
15	19.82	545	452	750	836
20	<u>20.09</u>	<u>545</u>	<u>453</u>	<u>750</u>	<u>818</u>
B-2 ^a	19.93	545	451	750	830

^aAverage values used for series B-2 are shown for comparison.

Table 2. Variation of droplet size and feed and product concentrations for series B-2

Run No.	Droplet size mm.	Concentration gm. HAC/liter			
		Aqueous feed	Raffinate	Organic feed	Extract
1	1.78	204.9	76.4	6.46	98.2
12	1.79	204.2	74.6	6.13	97.6
15	1.90	204.2	74.9	6.13	97.8
20	<u>1.70</u>	<u>204.7</u>	<u>76.2</u>	<u>6.34</u>	<u>96.7</u>
B-2 ^a	1.79	204.5	75.5	6.27	97.6

^aAverage values used for series B-2 are shown for comparison.

The internal column concentrations reported in Appendix C for groups A, B, C, and E represent smoothed data. Typical variations of these concentrations (11) from recorded smoothed data points are presented in Table 3.

Table 3. Variation of internal column concentration data for series B-2

Run No.	Concentration, gm. HAC/liter						
	x_{0u}	x_{1u}	y_{1u}	x_{2d}	x_{1d}	y_{1d}	y_{2d}
1	163.8	124.5	71.5	--	--	--	--
12 ^a	--	--	--	--	--	--	--
15	163.8	124.5	--	93.7	128.6	81.1	51.5
20	<u>163.0</u>	<u>125.9</u>	--	<u>91.0</u>	<u>128.7</u>	<u>79.0</u>	<u>49.4</u>
B-2 ^b	164.4	125.4	70.6	96.4	128.2	79.4	51.7

^aHoldup run.

^bSmoothed values used for series B-2 are shown for comparison.

In the interest of clarity, the operating variables for this study are summarized in Table 4. The over-all mass transfer coefficients based on the aqueous phase concentration gradient are tabulated in Table 5. The method of calculating these coefficients is shown by a sample calculation of series B-2 in Appendix B. The results for groups A, B, C, D, and E are represented graphically in Figures 8 through 12.

Table 4. Summary of operating variables

Group	No. of stages	Pulse frequency cpm.	Aqueous feed concentrations gm. HAC/liter	Aqueous feed rate ml./min.
A ^a	1	variable	204	545
B ^a	2	variable	204	545
C ^a	2	19.9	204	variable
D ^a	2	19.9	variable	499-545 ^b
E ^a	5	19.87	205.9	484
F ^c	2	16.03	204.5	200
G ^c	2	variable	207.6	191
H ^c	2	variable	106.5	194

^aMethyl isobutyl ketone-acetic acid-water system, pulse amplitude = 1.252 cm.

^bAqueous feed rate for series D-1 = 545, D-2 = 499, and D-3 = 542.

^cIsophorone-acetic acid-water system, pulse amplitude = 2.553 cm.

Table 5. Over-all mass transfer coefficients

Series code	Group variable	Amount transferred g. HAC	Driving force	Area ² Cm.	Average time sec.		Mass transfer coefficient ft./hr.
			<u>g. HAC</u> ml.		<u>Rise</u> sec.	<u>Formation</u> sec.	
Pulse frequency cpm.							
A-1	14.92	1.952	0.0377	1570	0.71	0.20	4.30
A-2	19.86	1.337	0.0316	1220	0.88	0.13	4.04
A-3	25.06	1.045	0.0316	1070	1.04	0.08	3.25
A-4	29.91	0.870	0.0344	828	0.93	0.09	3.41
A-5	35.27	0.748	0.0356	656	0.89	0.10	3.81
Aqueous feed rate ml./min.							
B-1	14.87	1.400	0.0346	1620	0.72	0.17	3.31
B-2	19.93	0.738	0.0259	1350	0.95	0.10	2.36
B-3	25.16	0.590	0.0193	1110	1.03	0.07	2.95
B-4	30.00	0.485	0.0139	949	1.06	0.06	3.87
C-1	152	0.338	0.0124	1190	0.88	0.12	2.69
C-2	348	0.564	0.0189	1290	0.93	0.10	2.66
C-3	545	0.738	0.0259	1350	0.95	0.10	2.36
C-4	742	0.961	0.0310	1390	0.95	0.10	2.49

Table 5. (Continued)

Series code	Group variable	Amount transferred g. HAC	Driving force <u>g. HAC</u> ml.	Area <u>cm.²</u>	Average time sec.		Mass transfer coefficient ft./hr.
					Rise sec.	Formation sec.	
Concentration aqueous feed weight %							
D-1	20.0	0.738	0.0259	1350	0.95	0.10	2.36
D-2	16.1	0.508	0.0218	1280	0.92	0.12	2.07
D-3	9.00	0.234	0.0126	1250	0.93	0.12	1.67
Stage No.							
E-1	1	1.013	0.0130	1710	0.80	0.10	6.00
E-2	2	0.626	0.0112	1630	0.81	0.11	4.40
E-3	3	0.353	0.0103	1410	0.72	0.16	3.25
E-4	4	0.213	0.0108	1290	0.68	0.21	2.03
E-5	5	0.206	0.0121	1370	0.70	0.17	1.67
Pulse frequency cpm.							
F-1	16.03	0.732	0.0225	1290	1.50	0.10	1.87
G-1	15.02	0.746	0.0186	1430	1.58	0.10	1.97
G-2	19.69	0.221	0.0156	1210	1.78	0.10	0.72
H-1	14.74	0.527	0.0102	1370	1.43	0.10	2.92
H-2	19.71	0.175	0.0105	1110	1.55	0.10	1.08

Figure 8. Effect of frequency on the mass transfer coefficient for the methyl isobutyl ketone-acetic acid-water system

Figure 9. Effect of frequency on the active interfacial area for the methyl isobutyl ketone-acetic acid-water system

Figure 10. Effect of aqueous feed rate on the mass transfer coefficient and active interfacial area for the methyl isobutyl ketone-acetic acid-water system

Figure 11. Effect of aqueous feed concentration on the mass transfer coefficient for the methyl isobutyl ketone-acetic acid-water system

Figure 12. Mass transfer coefficients and active interfacial area in a five stage column with the methyl iso-butyl ketone-acetic acid-water system

DISCUSSION OF RESULTS

The mass transfer coefficients represented graphically in Figure 8 as functions of pulse frequency exhibit minima. Before discussing the effects of frequency, however, the effect of concentration on the mass transfer coefficient will be considered.

The aqueous flow rate of series D-2 was about 10 percent lower than the aqueous flow rates of group D. The effect of the aqueous flow rate on the mass transfer coefficient was very slight; therefore, series D-2 was also included in this group. Figure 11 displays an increase of the mass transfer coefficient with increased concentration of acetic acid in the aqueous feed. As the acetic acid concentration in the aqueous feed was increased, the acid content in both the organic and aqueous phases became greater at every point in the column. The two phases consequently are becoming physically more similar. Moreover, the interfacial tension between the phases is reduced. The reduction in interfacial tension is very likely the cause for the increase in the mass transfer coefficient. This supports the postulate of Coulson and Skinner (5), who investigated mass transfer to single droplets for the methyl isobutyl ketone-benzoic acid-water and benzene-benzoic acid-water systems. Their experimental value for the mass transfer coefficient in the ketone system was three times those evaluated in the benzene system. They postulated that the

large difference in interfacial tension, 8.8 dynes/cm. for ketone-water and 35 dynes/cm. for benzene-water, may be the cause of the big difference in the mass transfer coefficients for the two systems.

A significant decrease in the mass transfer coefficient from the top to the bottom of the five stage column is indicated by Figure 12. The interfacial tension was the lowest at the top of the column because the aqueous feed which was rich in acetic acid was fed into the top. The observance of larger mass transfer coefficients at the top of the column where the interfacial tension was reduced strengthens the validity of the previously discussed effect of concentration.

Chu, et al. (3) stated that extraction efficiency was greatly enhanced by the lowering of interfacial tension by the addition of wetting agents. The performance of a packed column with benzene-benzoic acid-water as the system was reported using the H.T.U. concept. They assumed that adsorption of the wetting agents at the interface offered an additional film resistance to diffusion of the solute from one phase to another, as well as an increased interfacial area. The addition of the wetting agents Aerosol OT., Naccolene F., and Santomerse D. improved the rate of extraction, expressed as H.T.U. It was concluded that the factor determining the rate of diffusion when these three wetting agents were added was the increased interfacial area. The packing was able to break

up the dispersed phase into smaller droplets when the interfacial tension was reduced. This conclusion was made by a visual observation because Chu and his associates did not obtain a measured interfacial area in the packed column that they used. Addition of diethylene glycol dilaurate decreased the extraction efficiency by hindering the diffusion of the solute by the presence of molecules of the wetting agent at the interface. Although the method of varying the interfacial tension used by these investigators was a pertinent consideration, inability to conclusively compare the results of the foregoing study and the present study is perhaps explained by the difference in the visual procedure and the photographic technique of measuring free rising dispersed droplets.

In general, the mass transfer coefficients for the isophorone system were lower than those in the ketone system. Since the interfacial tensions for the two systems were nearly the same for the operating conditions used, the probable reason for the lower coefficients in the isophorone system was the high viscosity of the isophorone phase. In extraction, the film resistance in the dispersed droplet is relatively unaffected by any change in operating conditions. Thus to increase the extraction efficiency the resistance of the continuous film must be decreased; but this can be done only to a point and is then limited by the presence of the resistance in the dispersed phase droplet. The viscosity of the

organic phase thus was the important factor for the rate of mass transfer because the organic phase was the principal dispersed phase in both systems.

In the isophorone system, the effect of increased acetic acid concentration in the aqueous phase did not increase the mass transfer coefficient. However, the density difference in the isophorone system was about threefold less than that in the ketone system. This required drastic changes in pulse operating characteristics and thus, the discrepancies between the two systems were caused in part by the difference in these operating variables.

Over-all mass transfer coefficients were reported for the methyl isobutyl ketone-acetic acid-water system in a specialized laboratory model of a spray column by Licht and Conway (13). They reported values of K which ranged from 0.8 to 1.2 ft./hr. The values of K in this study ranged from 1.7 to 4.3 ft./hr. The dispersed phase in the study by Licht and Conway was an aqueous phase of 3 to 6 weight percent acetic acid, which made the viscosity of the dispersed aqueous phase 30 percent larger in their work than that of the dispersed organic phase used in this study. Also the interfacial tension in their dilute system of 3 to 6 weight percent acetic acid in water was 50 percent greater than that used in this study. Therefore, the difference in the values of K between the two studies is reasonable.

On the basis of concentration effects on the mass transfer coefficients, we can now explain the effect of increasing the pulse frequency. In groups A and B, the pulse frequency of 15 cycles per minute corresponded to a condition of operation near the lower flooding limit, which closely approximates mixer settler operation. Little or no recycling or backmixing of the aqueous phase occurs at the lower flooding limit. Thus the absence of recycling of the aqueous phase produces a higher acetic acid concentration in both phases corresponding to a low interfacial tension, which in turn will yield a higher mass transfer coefficient. As the pulse frequency was increased, the acetic acid concentration in stage 1 was reduced by recycle or backmixing of the aqueous phase and the mass transfer coefficient was decreased. Further increasing of the pulse frequency from 20 or 25 cycles to 30 or 35 cycles does not reduce the acetic acid concentration as much as that observed near the lower flooding limit, even though the amount of aqueous recycle stream is greater than the organic stream pushed upward through the column. The dilution of the aqueous phase is overcome by the greater turbulence to both phases at the higher pulse frequencies. The resistance of the continuous phase film is now reduced and the mass transfer coefficient becomes larger. It is of interest to note that the up-stroke efficiency reported by Burkhardt and Fahien (2) also exhibits a minimum when represented as a function of pulse

frequency.

The active interfacial area for mass transfer was based on the organic phase which rose freely through the stage. The active area decreased with increasing pulse frequency, even though the diameter of a droplet of average specific surface became smaller, because of the amount of organic phase pushed upward through the column was less per cycle with increased frequency. The decreased area is shown in Figure 9.

The mass transfer coefficients for group C are represented graphically as a function of aqueous feed rate in Figure 10. The line was determined by the method of least squares. The aqueous feed concentration and pulse frequency were held constant in each series in this group; one would expect the mass transfer coefficient to increase with increased aqueous feed rate because of the greater acetic acid concentration at the higher aqueous feed rates. However, the opposite effect was observed. It is postulated that if the aqueous feed concentration were adjusted to maintain a constant aqueous feed concentration in the stage under consideration that the mass transfer coefficients at the lower aqueous feed rates would be even greater and thus the effect of increasing the aqueous feed rate would greatly decrease the mass transfer coefficient. The ratio of organic flow rate to aqueous flow rate possibly would be a better variable. This is supported by the work in the 5 stage column (group E) where

the ratio of the organic to aqueous flow rate was approximately 2.3, which was the greatest ratio of organic to aqueous flow rate used in the study. The mass transfer coefficients for the top two stages of the column were the largest observed.

NOMENCLATURE

- A = cross sectional area of column, cm.²
a = pulse amplitude, cm.
B = organic feed rate, ml./cycle.
 d_{vs} = diameter of a drop of average specific surface, cm.
E = extract flow rate, ml./cycle.
F = aqueous feed rate, ml./cycle.
f = pulse frequency, cycles/min.
G = internal organic flow rate, ml./cycle.
H = holdup of organic (dispersed) phase, cm.³/stage.
K = mass transfer coefficient, cm./hr.
L = internal aqueous flow rate, ml./cycle.
N = amount of mass transferred, gms. HAc.
R = raffinate flow rate, ml./cycle.
S = active interfacial area, cm.²
t = time of mass transfer, sec.
V = volume of stage, cm.³ (with no subscript); volume of aqueous phase recycle, ml./cycle (with subscript R).
x = concentration of solute in aqueous phase, gm./ml.
y = concentration of solute in organic phase, gm./ml.
 ΔC_{lm} = logarithmic mean driving force based on aqueous phase concentration units, gm./ml.

Subscripts

- B = organic feed
d = end of downstroke

E = extract

F = aqueous feed

R = recycle with V only; equals raffinate with all others.

t = transient condition.

u = upstroke

0 = stage 0 above top plate.

1 = stage 1.

2 = stage 2.

* = equilibrium condition.

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APPENDIX A

System Properties

The equilibrium data and physical properties of the methyl isobutyl ketone-acetic acid-water system and isophorone-acetic acid-water system were tabulated in Tables 6 and 7. The viscosity data were taken with an Ostwald viscometer. The interfacial tension data were obtained by the droplet-weight-volume method.

The equilibrium data and physical properties of the methyl-isobutyl ketone-acetic acid-water system agree well with those reported by Honekamp (9) and Karr and Scheibel (10).

The equilibrium data for the isophorone-acetic acid-water system compares favorably with data reported by Othmer, et al. (14). The interfacial tension at high acetic acid concentrations was not obtained experimentally for this system because the aqueous phase would not wet the burette tip properly. The values were taken from an extrapolated curve of interfacial tension as a function of organic weight percent.

Table 6. Equilibrium data and physical properties of the methyl isobutyl ketone-acetic acid-water system at 25° C

Concentration gm./liter		Weight percent		Density gm./liter		Viscosity centipoise		Inter- facial tension
organic	aqueous	organic	aqueous	organic	aqueous	organic	aqueous	dynes/cm.
9.16	17.74	1.139	1.778	0.80410	0.99790	0.605	0.974	9.967
17.25	32.87	2.136	3.287	0.80746	1.0001	0.618	1.000	9.357
26.41	48.81	3.256	4.87	0.81119	1.0021	0.639	1.030	8.444
34.77	62.68	4.272	6.18	0.81395	1.0039	0.660	1.058	8.001
43.98	75.43	5.38	7.50	0.81682	1.0054	0.680	1.083	7.158
70.08	113.6	8.48	11.26	0.82631	1.0093	0.756	1.166	5.683
89.59	139.2	10.74	13.75	0.83381	1.0121	0.808	1.220	4.780
117.3	173.7	13.89	17.11	0.84415	1.0152	0.894	1.301	3.464
141.2	201.1	16.54	19.76	0.85372	1.0173	0.975	1.367	2.863
168.1	229.2	19.46	22.50	0.86381	1.0189	1.068	1.439	2.059

Table 7. Equilibrium data and physical properties of the isophorone-acetic acid-water system at 25.5° C

Concentration gm./liter		Weight percent		Density gm./liter		Viscosity centipoise		Inter- facial tension dynes/cm.
organic	aqueous	organic	aqueous	organic	aqueous	organic	aqueous	
0.00	0.00	0.00	0.00	0.92054	0.99709	2.375	0.925	8.05
64.2	60.47	6.87	6.02	0.93489	1.0052	2.865	1.037	5.61
125.2	121.5	13.21	11.99	0.94784	1.0131	3.222	1.159	3.59
170.8	170.8	17.85	16.78	0.95706	1.0181	3.347	1.282	2.23
218.1	220.6	22.54	21.57	0.96768	1.0225	3.383	1.427	1.35
260.1	226.8	26.58	26.03	0.97844	1.0249	3.350	1.632	0.70 ^a
301.6	307.9	30.37	30.11	0.99315	1.0227	3.237	2.042	0.10 ^a

^aExtrapolated values.

APPENDIX B

A typical calculation for mass transfer coefficient is shown for series B-2, C-3, or D-1. These data are identical, only the code has been changed to keep the lettering within a group of data consistent.

The necessary data for this calculation are:

Flow rates

Aqueous feed	$F = 27.33 \text{ ml./cycle}$
Organic feed	$B = 37.64 \text{ ml./cycle}$
Extract	$E = 41.65 \text{ ml./cycle}$
Raffinate	$R = 22.62 \text{ ml./cycle}$

Aqueous concentrations

Aqueous feed	$x_F = 0.2045 \text{ gm./ml.}$
Raffinate	$x_R = 0.0755 \text{ gm./ml.}$
End of upstroke in stage 0	$x_{0u} = 0.1644 \text{ gm./ml.}$
End of upstroke in stage 1	$x_{1u} = 0.1254 \text{ gm./ml.}$
End of downstroke in stage 1	$x_{1d} = 0.1282 \text{ gm./ml.}$
End of downstroke in stage 2	$x_{2d} = 0.0964 \text{ gm./ml.}$

Organic concentration

Organic feed	$y_B = 0.00627 \text{ gm./ml.}$
Extract	$y_E = 0.0976 \text{ gm./ml.}$
End of upstroke in stage 1	$y_{1u} = 0.0706 \text{ gm./ml.}$
End of downstroke in stage 1	$y_{1d} = 0.0794 \text{ gm./ml.}$
End of downstroke in stage 2	$y_{2d} = 0.0517 \text{ gm./ml.}$

Other data

Pulse amplitude	$a = 1.252 \text{ cm./cycle}$
Pulse frequency	$f = 19.93 \text{ cycles/min.}$
Cross sectional area of column	$A = 36.87 \text{ cm.}^2$
Holdup of organic in Stage 1	$H = 52.30 \text{ cm.}^3$
Total volume of Stage 1	$V = 283.9 \text{ cm.}^3$
Diameter of a droplet of average specific surface	$d_{vs} = 0.179 \text{ cm.}$

Calculation of Internal Flow Rates

The pulse column internal flow rates were calculated from the equation derived by Edwards (6) and modified for continuous raffinate removal by Graham (7). This equation on a per cycle basis for the top of Stage 1 becomes,

$$V_{R_1} = aA \cosh \frac{G_1 - L_0}{\tau aA} - \frac{1}{2} (G_1 + L_0) \quad (1)$$

These terms are explained above and in Figure 13.

The organic flow rate out of a stage was assumed to be proportional to the solute concentration in this stream, or for Stage 1,

$$\begin{aligned}
 G_1 &= E - (E-B) \frac{(y_E - y_{1d})}{(y_E - y_B)} \\
 &= 41.65 - (41.65) \frac{(0.0976 - 0.0794)}{(0.0976 - 0.00627)} \\
 &= 40.86 \text{ ml./cycle.}
 \end{aligned}$$

The quantity $G_1 - L_0$ was taken as the average of the

Figure 13. Internal concentrations and flow rates in a pulse column

extract rate minus the aqueous feed rate at the top of the column and the organic feed rate minus the raffinate rate at the bottom on the column.

$$\begin{aligned} G_1 - L_0 &= \frac{1}{2} [(E - F) + (B - R)] \\ &= \frac{1}{2} [(41.65 - 27.33) + (37.64 - 22.62)] \\ &= 14.67 \text{ ml./cycle} \end{aligned}$$

$$\therefore L_0 = G_1 - (G_1 - L_0) = 40.86 - 14.67 = 26.19 \text{ ml./cycle.}$$

The amount of aqueous recycle becomes, by Equation 1,

$$\begin{aligned} V_{R_1} &= (1.252)(36.87) \cosh \frac{14.87}{(3.142)(1.252)(36.87)} \\ &\quad - \frac{1}{2} (40.86 + 26.19) \\ &= 12.88 \text{ ml./cycle} \end{aligned}$$

and

$$(L_0 + V_{R_1}) = 26.19 + 12.88 = 39.07 \text{ ml./cycle.}$$

Internal flow rates $G_2 = 39.63 \text{ ml./cycle}$, $V_{R_2} = 14.11 \text{ ml./cycle}$, and $(L_1 + V_{R_2}) = 39.07 \text{ ml./cycle}$ are calculated in the same manner.

Calculation of Internal Concentrations

Negative driving forces were encountered in preliminary calculations when all experimentally determined internal concentrations were used. Therefore, the three concentrations, Y_{1d} , Y_{2d} , and x_{1d} , were calculated. Both of the measured and calculated values are reported in Appendix C.

At steady state the continuous upflow or downflow of acetic acid must be the same at any point in the column. For this series the average downflow of acetic acid per cycle was

$$\begin{aligned} & \frac{1}{2}[(Fx_F - Ey_E) + (Rx_R - By_B)] \\ &= \frac{1}{2}[(27.33)(0.2045) - (41.65)(0.0976) + (22.62)(0.0755) \\ & \quad - (37.64)(0.00627)] \\ &= 1.500 \text{ gm. HAC downward/cycle.} \end{aligned}$$

Now y_{2d} can be solved directly by an acetic acid balance at plate 2.

$$(L_1 + V_{R_2}) x_{1u} - 1.500 = G_2 y_{2d} + V_{R_2} x_{2d}$$

$$(39.07)(0.1254) - 1.500 = 39.63 y_{2d} + (14.11)(0.0964)$$

$$y_{2d} = 0.0515 \text{ gm./ml.}$$

Similarly, the acetic acid balance at plate 1 becomes,

$$(L_0 + V_{R_1}) x_{0u} - 1.500 = G_1 y_{1d} + V_{R_1} x_{1d}$$

$$(39.07)(0.1644) - 1.500 = 40.86 y_{1d} + 12.88 x_{1d}$$

$$4.923 = 40.86 y_{1d} + 12.88 x_{2d} \quad (2)$$

The grams of acetic acid transferred to the organic phase from the aqueous phase was

$$N = Hy_{1u} - (H - G_1)y_{1d} - G_2 y_{2d}$$

where N = amount of solute transferred during an upstroke, gm.

Thus,

$$N = (52.30)(0.0706) - (52.30 - 40.86)y_{1d} - (39.63)(0.0515)$$

$$N = 1.653 - 11.44 y_{1d} \quad (3)$$

Also, the amount of acetic acid transferred from the aqueous phase to the organic phase was

$$N = (V - H - V_{R1})x_{1d} + V_{R2} x_{2d} - (V - H)x_{1u}$$

$$N = (283.9 - 52.30 - 12.88)x_{1d} + (14.11)(0.0964) - (283.9 - 52.30)(0.1254)$$

$$N = 218.7 x_{1d} - 27.68 \quad (4)$$

Solving Equations 2, 3, and 4 simultaneously

$$x_{1d} = 0.1293$$

$$y_{1d} = 0.0800$$

$$N = 0.738 \text{ gms. HAC transferred during upstroke.}$$

Calculation of Driving Force

Because of the countercurrent nature of the pulse column, the driving force based on the aqueous phase was calculated by the following equation,

$$\Delta C_{lm} = \frac{(x_{1d} - x_{2d}^*) - (x_{1u} - x_{1t}^*)}{2.303 \log \frac{(x_{1d} - x_{2d}^*)}{(x_{1u} - x_{1t}^*)}} \quad (5)$$

where x_{2d}^* = concentration of the aqueous phase in equilibrium with y_{2d} in gm. HAC/ml.

y_{1t}^* = concentration of the aqueous phase in equilibrium with y_{1t} in gm. HAC/ml. y_{1t} is the transient

acetic acid concentration in the organic phase, G_2 , from stage 2 after picking up N grams of acetic acid but before coalescence with organic phase, $H - G_1$, still remaining in stage 1.

In calculating this transient concentration, it was assumed that the volume of this organic phase was equal to G_1 since most of the acetic acid transferred to the organic phase takes place during the upstroke.

$$\text{Thus } y_{1_t} = \frac{G_2 y_{2_d} + N}{G_1}$$

$$y_{1_t} = \frac{(39.63)(0.0515) + 0.738}{40.86} = 0.068 \text{ gm./ml.}$$

This is in equilibrium with $x_{1_t}^* = 0.1110 \text{ gm./ml.}$ and $y_{2_d} = 0.0515 \text{ gm./ml.}$ is in equilibrium with $x_{2_d}^* = 0.0870 \text{ gm./ml.}$

Therefore, using Equation 5

$$\Delta C_{lm} = \frac{(0.1293 - 0.0870) - (0.1254 - 0.1110)}{2.303 \log \frac{0.0423}{0.0144}}$$

$$\Delta C_{lm} = 0.0259 \text{ gm. HAc/ml.}$$

Calculation of Interfacial Area

The active interfacial area for mass transfer was based on the arithmetic average volume of organic phase which rose freely through stage 1 during an upstroke. The remaining organic phase in stage 1, $H - G_1$, was for the most part in the

coalesced form during the upstroke. Those droplets which were not coalesced were very close together and their available area was shielded greatly. Thus, the volume of organic phase which contributes to the active interfacial area was $\frac{1}{2} (G_1 + G_2)$.

The volume of a droplet with diameter d_{vs} is $\frac{1}{6} \pi d_{vs}^3$ and the surface area is πd_{vs}^2 . Therefore, the active interfacial area becomes,

$$S = \frac{\frac{1}{2} (G_1 + G_2) \pi d_{vs}^2}{\frac{1}{6} \pi d_{vs}^3} = \frac{3 (G_1 + G_2)}{d_{vs}}$$

$$= \frac{3 (40.86 + 39.63)}{0.179} = 1350 \text{ cm.}^2$$

Mass Transfer Coefficient Calculation

The mass transfer coefficients were calculated using the equation

$$\frac{N}{t} = K S \Delta C_{lm} \quad (6)$$

This equation is written assuming equilibrium conditions exist at the interface and that the equilibrium relation is a straight line over the range of concentrations in the stage.

Equation 6 also assumes a steady state area. Since the area in a pulse column is formed and collapses during each cycle, an evaluation of the transfer time and interfacial area relationship must be made.

If all the droplets formed during an upstroke of the pulse generator are assumed to be of the same diameter, d_{vg} , the average time of formation of this droplet and the rise time through the stage based on the terminal velocity can be calculated. The time of formation plus the rise time is the transfer time for one droplet, assuming that the time for this droplet to reach terminal velocity is negligible. Since all droplets are assumed to have the same diameter, d_{vg} , the transfer time for each droplet will be the same. Even though the droplets are not all formed at the same time, the total active interfacial area, however, is exposed to the continuous phase for a period of time equal to the transfer time for only one droplet.

The rise time for droplets of diameter, d_{vg} , was calculated by a method proposed by Coulson and Richardson (4). This calculation is based on the assumption that the droplets are rigid spheres. The calculation of the rise time and the average formation time is also summarized in the study by Graham (7).

The calculated rise time for a droplet of diameter, d_{vg} , for series B-2 was 0.95 sec. The average formation time was 0.10 sec. while the time for this droplet to reach terminal velocity was only 0.015 sec. This justifies the previous assumption.

Thus

$$t = 0.95 + 0.10$$

$$= 1.05 \text{ sec.}$$

and

$$K = \frac{N}{S \Delta C_{lm} t}$$

$$= \frac{(0.738) (3600)}{(1350) (0.0259) (1.05) (30.48)}$$

$$= 2.36 \text{ ft./hr.}$$

APPENDIX C**Experimental Data**

Table 8. Calculated and experimental data

Code No.	Low rates, ml./min.				Pulse frequency cpm.	Organic holdup cm. ³ /stage	d_{vs} mm.	Calculated concentrations		
	Aqueous feed	Raffinate	Organic feed	Extract				gm. HAC/liter x_{1d}	y_{1d}	y_{2d}
Methyl isobutyl ketone-acetic acid-water system ^a										
A-1	545	464	750	824	14.92	83.94	2.04	126.1	59.3	31.3
A-2	545	459	750	840	19.86	46.11	1.98	122.0	62.4	31.4
A-3	545	463	750	832	25.06	33.42	1.75	120.9	63.9	31.6
A-4	545	468	750	817	29.91	30.12	1.92	121.3	64.9	30.8
A-5	545	471	750	813	35.27	31.89	2.05	119.7	66.9	28.5
B-1	545	542	750	828	14.87	86.46	2.00	141.0	72.3	48.2
B-2	545	451	750	830	19.93	52.34	1.79	129.3	80.0	51.3
B-3	545	449	750	840	25.16	42.70	1.75	125.7	80.2	52.8
B-4	545	448	750	837	30.00	39.00	1.71	123.7	80.5	54.4
C-1	152	111	750	781	19.86	37.83	1.94	52.5	28.8	15.6
C-2	348	276	750	817	19.86	43.41	1.85	94.7	58.0	34.6
C-3	545	451	750	830	19.93	52.34	1.79	129.3	80.0	51.3
C-4	742	649	750	840	19.86	61.38	1.77	155.0	88.9	62.7
D-1	545	451	750	830	19.93	52.34	1.79	129.3	80.0	51.3
D-2	499	432	750	804	19.85	43.81	1.86	102.1	59.6	39.0
D-3	542	515	750	765	19.88	41.48	1.83	65.7	36.3	25.8
E-1	484	372	1000	1090	19.87	81.22	1.88	115.7	66.6	46.3
E-2	484	372	1000	1090	19.87	64.50	1.93	81.8	46.4	31.9
E-3	484	372	1000	1090	19.87	57.96	2.20	55.5	31.9	20.8

^aThe value of pulse frequency cpm. for the methyl isobutyl ketone-acetic acid-water system was 1.252.

Table 8. (Continued)

Code No.	Low rates, ml./min.				Pulse frequency cpm.	Organic holdup cm. ³ /stage	d _{vs} mm.	Calculated concentrations		
	Aqueous feed	Raffinate	Organic feed	Extract				x _{1d}	y _{1d}	y _{2d}
E-4	484	372	1000	1090	19.87	58.80	2.38	39.5	19.96	12.7
E-5	484	372	1000	1090	19.87	54.83	2.23	29.6	12.7	7.7
Isophorone-acetic acid-water system ^b										
F-1	200	130	650	713	16.03	52.78	2.00	58.0	46.9	25.6
G-1	191	142	650	700	15.02	51.34	1.91	56.6	48.0	27.5
G-2	191	143	650	686	19.69	53.35	1.69	57.5	54.2	34.9
H-1	194	165	650	674	14.74	50.65	1.97	33.2	22.8	15.7
H-2	194	172	650	656	19.71	48.95	1.80	32.3	32.9	18.9

^bThe value of pulse amplitude for the isophorone-acetic acid-water system measured in cm./cy. was 2.553.

Table 9. Acetic acid concentration data

Code No.	Measured concentrations gm. HAc/liter										
	x_F	x_R	y_B	y_E	x_{0u}	x_{1u}	y_{1u}	x_{2d}	x_{1d}	y_{1d}	x_{2d}
A-1	204.7	93.4	6.57	88.9	165.0	124.8	64.0	93.5	127.0	66.2	37.2
A-2	204.5	92.4	6.46	88.0	154.9	115.2	62.6	92.5	116.7	68.1	19.2
A-3	204.5	99.3	6.35	84.4	149.8	115.5	62.5	99.2	117.0	69.7	40.1
A-4	204.6	104.2	6.57	83.0	146.9	116.6	62.3	104.4	117.8	71.0	42.1
A-5	204.0	107.1	6.49	81.4	144.6	116.0	63.0	107.1	116.8	71.6	44.0
B-1	204.7	76.4	6.39	97.1	169.2	134.5	72.2	104.7	137.5	77.3	49.0
B-2	204.5	75.5	6.27	97.6	164.4	125.4	70.6	96.4	128.2	79.4	51.7
B-3	204.7	81.2	6.37	94.9	157.0	121.2	71.7	95.2	125.2	78.0	54.0
B-4	204.7	83.3	6.34	92.1	151.6	119.3	73.6	96.0	124.5	78.6	55.7
C-1	203.9	30.5	6.47	40.7	78.5	50.2	24.0	35.9	52.5	28.8	15.4
C-2	203.3	55.0	6.46	73.6	131.9	90.4	49.4	65.5	93.9	55.2	14.7
C-3	204.8	75.5	6.27	97.6	164.4	125.4	70.6	96.4	128.2	79.4	51.7
C-4	203.4	100.2	6.47	108.8	178.5	150.3	85.5	125.0	152.7	98.2	54.0
D-1	204.8	75.5	6.27	97.6	164.4	125.4	70.6	96.4	128.2	79.4	51.7
D-2	163.1	63.0	6.40	72.8	129.1	98.66	52.4	75.24	102.3	58.91	14.7
D-3	90.92	43.3	6.40	40.9	78.48	64.01	31.89	52.42	64.79	34.89	24.0
E-1	205.9	24.6	5.41	87.6	152.2	109.7	65.0	81.81	115.0	70.4	42.0
E-2	205.9	24.6	5.41	87.6	109.7	78.0	43.8	55.53	80.6	48.0	15.4
E-3	205.9	24.6	5.41	87.6	78.0	53.3	27.1	39.5	56.1	34.72	24.0
E-4	205.9	24.6	5.41	87.6	53.3	38.1	17.2	29.6	40.7	20.0	24.0
E-5	205.9	24.6	5.41	87.6	38.1	28.5	11.8	24.92	30.0	13.1	24.0
F-1	204.5	34.5	6.61	56.2	76.0	50.32	24.16	41.37	64.49	50.32	15.4
G-1	207.6	33.9	8.17	57.6	75.66	48.43	43.72	38.49	61.61	43.9	24.0
G-2	207.6	35.1	8.17	57.8	73.92	50.68	48.16	38.42	61.67	47.8	24.0
H-1	106.5	21.1	7.33	32.4	40.05	28.52	26.72	24.32	36.09	26.3	15.4
H-2	106.5	21.9	7.33	33.01	41.24	29.33	26.96	25.16	36.45	27.38	15.4