MIXING IN A FLUID FLOWING THROUGH A PACKED BED

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May 26, 1947

Professor J. S. Newell
Secretary of the Faculty
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Dear Sir:

In accordance with the regulations of the Faculty, we hereby submit a thesis entitled "Mixing in a fluid Flowing Through a Packed Bed," in partial fulfillment of the requirements for the degree of Master of Science in Chemical Engineering Practice.

Very sincerely yours,

A. C. Sugden

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ACKNOWLEDGEMENT.

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I SUMMARY.

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The mixing processes which occur in a liquid flowing through a packed tower have been studied and measured.

Water was passed up a packed tower, and an indicator solution of methylene blue injected at the axis. The rate at which mixing occurred was determined by taking samples from various points and measuring their concentrations. The process of mixing was found to conform to the normal diffusion law, or to the law derived from "random walk" theory. (the two give very nearly the same expression), and an "apparent diffusion coefficient" D could be calculated from the distribution.

For spherical particles (with which most of the work was done):

$$\frac{D\rho}{\mu} = 0.035(Re)^{1.4}$$

(where (Re) is the modified Reynolds' number), when (Re) was in the range 10 - 100.

In this range it is considered that flow is turbulent in some parts of the packing, and streamline in others. The mixing process is thought to be caused by:

local turbulence (leading to "eddy diffusion) : splitting and scattering of streams by channeling (caused by fluctuations in the specific resistance to flow of various parts of the packing) : and possibly by displacement of elements of the liquid when they impinge on the particles.

II INTRODUCTION.

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Utilitarian aspect of the problem. At the moment fixed-bed catalytic reactors are designed largely on the "hit-or-miss" or "build-and-try" principle, simply because it is not possible to calculate the dimensions and other characteristics of a reactor from small-scale experiments. Since the rate of a gaseous reaction at any point in the catalyst bed depends on the temperature and composition of the gas, it is clear that the rationalisation of reactor design must depend on a knowledge of the coefficients of heat and mass transfer through the bed. As regards the latter, it is known that the molecules of the gas do not in general travel in straight lines parallel to the apparent axis of flow, but follow a devious path, so that mixing takes place transversely and longitudinally. The temperature and composition of the gas at any point will, therefore, depend, among other things, on the rate at which this mixing takes place.

Object of Present Work. The experimental work described in this thesis was designed to throw some light on the mechanism and quantitative characteristics of the mixing which occurs when a fluid flows through a bed of particles of uniform size and shape. Since liquids are easier to handle than gases, it was decided to do the preliminary work using water as the moving fluid, on the supposition that there would be a general similarity to the case of a gas-stream, particularly at comparable Reynold's numbers. Although it was desirable to cover the range of Reynolds' numbers which might be expected in a catalytic reactor, it was also of interest to gain a picture of the behavior of the system over as wide a range as possible. In practice, the upper limit of flow rate was fixed by the limitations of the apparatus, and the lower by limitations in the technique of measurement employed; the higher Reynolds' numbers were of the order of magnitude of those which might be encountered in a reactor.

Nature of the Phenomenon. In a gas-stream, mixing may be expected to occur in several ways during passage through a packing: normal diffusion, caused by thermal motion of the molecules: mixing due to turbulence: and possibly scattering caused by the diversion of various parts of the stream as they impinge on solid surfaces. In a liquid it was assumed (and with justification, as the experiments showed) that the normal diffusion coefficient would be negligibly small in comparison with the other effects at the rates of flow employed. We are thus left with turbulent mixing (or eddy-diffusion) and with the possibility of another distinguishable mechanism, which will be called displacement mixing.

Although no previous work on packed tubes had been published, a good deal has been done on eddy-diffusion in open tubes. Mixing occurs in a fluid in turbulent flow, because any element of fluid may have a backward, forward or sideways component of velocity superimposed on its mean flow velocity. The size of the "element" undergoing these fluctuations, the magnitude of the random velocities attained and the mean distance travelled between changes of direction are indeterminate; but the effective result is a mixing process, analogous to molecular diffusion, and susceptible to similar mathematical treatment.

In the case of molecular diffusion, the coefficient of diffusion D^1 is defined by the expression

$$\frac{dn}{dt} = A.D'.\frac{dc}{dx}$$

where $\frac{dn}{dt}$ is the rate at which molecules of the diffusing species cross area A of a plane at right-angles to the X-coordinate, c being the concentration of the diffusing species in molecules per unit volume. This is equivalent to the expression

$$\frac{\partial c}{\partial t} = A \cdot D' \frac{\partial^2 c}{\partial^2 x^2}$$

The foregoing expressions are true for the case where the concentration varies in one dimension only. For the more general case of variation in 3 dimensions:

$$\frac{\partial c}{\partial t} = A \cdot D' \left(\frac{\partial^2 c}{\partial x^2} + \frac{\partial^2 c}{\partial y^2} + \frac{\partial^2 c}{\partial z^2} \right)$$

The general validity of these diffusion laws can be demonstrated by considering the effects of random, independent movements of the diffusing molecules, such as occur in a gas. In a stagnant mass of gas, for instance, containing two or more species of molecule, any macroscopic variations in composition will in time disappear, because purely random movement will cause a net transfer of molecules of each species from regions where their concentration is high into regions where it is low. It is only necessary to assume that an individual molecule is equally likely to move in any direction at any moment, and that the mean length of individual displacements in all directions is equal to show that the diffusion laws given above will follow. (see Kennard's "Kinetic Theory of Gases") —— Subject to the limitations mentioned later.

In turbulent motion the elements undergoing random displacement are of macroscopic dimensions, and not individual molecules; the velocity of displacement will be comparable to the flow-velocity, and hence much slower than the thermal velocity in a gas; and the "mean free path" will no doubt be much greater than for molecular movements. It would be expected, however, that the net effect, in a system large enought to dwarf the fluctuations due to turbulence, would be analogous to the molecular diffusion.

The theory of eddy diffusion, which will not be discussed in detail here, has been developed largely by G. I. Taylor (10); the work of v. Karman (11), Prandtl (20), Dryden (12), Sherwood and Woertz (13), Murphree (14), Goldstein (15) may also be consulted. In place of the "mean free path" of kinetic theory the concept of "mixing length" is employed—the mixing length being the mean distance which an eddy travels before breaking up and losing its identity; and the root mean square velocity is replaced by the mean instantaneous deviating velocity in a direction perpendicular to the main flow. Neither of these quantities can readily be derived from the parametus of the system; in fact the present state of the theory does not admit of a priori predictions of the eddy diffusivity (or eddy diffusion coefficient) such as can be made for the molecular diffusivity D from kinetic theory.

However, experimental work has shown that the turbulent mixing process can be described by the normal mathematical diffusion laws, substituting an eddy-diffusivity E in place of D^1 — see Sherwood and Woertz (13), Sherwood and Towle (5), Weddell (17), Hawthorne (18), all of whom were investigating turbulence in open tubes, containing no packing. A similar conclusion was reached by Kalinske and Pien (19), who investigated mixing in open troughs.

When a fluid flows through a bed of solid particles at a sufficient rate to give rise to turbulence, therefore, it is to be expected that a diffusion-like mixing process will be caused by eddy-diffusion, but it is not possible to make quantitative predictions. At lower rates of flow, where turbulence is non-existent or negligible, it would appear doubtful at first sight whether mixing would occur at all; it might be expected that any two particles which entered the system close together would remain close together. This certainly is the case in a fluid in streamline flow past a single sphere (see Fig. IV(i). However, consideration of conditions in an assembly of spheres suggest that two particles which are separated by the splitting of the stream by a sphere may never regain their former relative positions, owning to the diversion of streams of fluid by other spheres in the assembly into the "gap" between the particles (see Fig. IV (ii)). Thus it is possible, without postulating discontinuities in the velocity distribution such as occur in turbulent motion, to conceive of purely streamline or viscous flow leading to permanent separation of particles which were originally close together. This is in effect a mixing process, and an attempt has been made to derive an expression for the corresponding mixing or diffusion coefficient, making certain simplifying assumptions (see Appendix "A"). This coefficient is referred to as the "geometrical" mixing coefficient, and the process as displacement mixing. However, considerations advanced in Appendix B suggest that the situation is complicated by the existence of channels in the packing, which lead to gross variations in the mass-velocity from region to region, so that the path of a particle is probably not determined solely or even predominantly by the configuration of the solids in its immediate neighbourhood.

The only experimental investigations of fluid mixing in a packed tower have been carried out by Rabinowitz and Roessel (4) , who worked out the apparatus and technique which have, with certain modifications, been used in the present work. It is thought, however, that their quantitative results were probably in error, owing to the excessive rate at which the indicator solution was introduced (see Appendix E).

<u>Diffusion and random scattering</u>. Rabinowitz and Roessel used an equation known as the Wilson equation (6) to calculate the mixing or apparent diffusion coefficient from measurements of concentration at various points in the tower. The Wilson equation is a solution of the general diffusion equation given earlier in this section, after the appropriate conditions have been inserted, and making the assumption that diffusion parallel to the axis of flow (i.e. longitudinal mixing) may be ignored. The present authors derived an equation similar to that used by Kalinske and Pien (19), by considering the distribution attained by particles undergoing random scattering; the derivation and final form of this "probability equation" is given in Appendix C. There is a discrepancy between the Wilson equation and the probability equation, although under the conditions of the experiments described here the divergence was within

the experimental error. The reaon for the discrepancy between the two equations is discussed in Appendix C.

III APPARATUS AND PROCEDURE.

III. Apparatus and Procedure (See Appendix E forfurther details)

Apparatus. This consisted of a constant head tank, providing a flow of water which was metered by an orifice and manometer. The water flowed up through a container filled with coarse packing, and then through a 4-inch glass tower which contained the packing under investigation (supported on wire gauze), and finally overflowed at the top of the tower. The indicator solution was introduced at the bottom of the tower by a capillary tube set on the axis. Samples could be withdrawn from various points on th e packing by means of five 1/64 inch steel capillary tubes inserted into the packing from above; these were rigidly mounted on a metal frame which held them vertically with their open ends in one horizontal plane and situated along a diameter of the tower. The samples were collected simultaneously in five test-tubes by applying a vacuum to the air-spaces of the latter. The tips of the sampling tubes were usually about an inch below the upper surface of the packing, and the total depth of packing in the tower was varied as convenient.

Procedure. Water was admitted at thedesired constant rate, and then the indicator solution (methylene blue, 25 gms./litre) was admitted through the injection tube. The rate of injection of indicator was adjusted to give a convenient depth of colour for the concentration determinations (see Appendix E). When steady state hed been reached a vacuum was applied to the collecting-tubes, and samples withdrawn from the tower. The first portions of liquid collected were thrown away, as they served to wash out the collecting-tubes. The samples proper were then collected. It appeared desirable to do this as slowly as possible, to avoid drawing into the tubes liquid from remote parts of the system. The time required to withdraw liquid through the capillary tubes at the same linear velocity as that of the stream through the packing would have been prohibitive, so the lowest convenient rate was used --10-20 minutes for a sample of about 25cc. from each tube--i.e. 2-20 times the linear flow velocity of the stream. The effect of this was to increase the effective area of cross section from which the sample was withdrawn; it is not believed to have distorted the results. It had been determined (see Appendix C)that the interpretation of the results did not depend on a constant rate of injection of indicator, as long as the mean concentration of the effluent was used for C* in the computation. Since the injection rate was not positively controlled, samples of effluent were taken at regular intervals during the sampling period, and mixed. There was a tendency for air to come out of solution and form bubbles on the packing, and it was feared that this might affect the results. Preheating of the water was tried and found to be useless. The effect was mitigated by stirring the packing before each run, thus dislodging the bubbles, which did not reach a size comparable with the packing particles during the course of a run.

Colorimetric analysis of methylene blue solutions. Methylene blue was used as an indicator because it can be estimated accurately and easily by its colour at great dilutions. Owing to the limitations on the rate of injection of indicator (discussed in Appendix E), the concentrations to be measured were as low as 1/4000 that of the indicator solution injected. The solution used, since it contained only 25 gms. of methylene blue to a litre, was considered unlikely to differ greatly in density or viscosity from pure water.

A Duboscq type colorimeter was used. The standard solution used for analysis was a $\frac{1}{1000}$ dilution of the indicator solution. A curve was constructed experimentally, showing color-density as a function of concentration (departure from Beer's law is great). A number of sub-standards of various concentrations were also used. To determine the concentration of a sample, its color-density was determined by comparison in the colorimeter with one of the standards (the nearest one, because color as well as density appeared to alter with concentration) and its concentration read off the curve.

Pressure-gradient Measurements. In order to measure the pressure-gradient through the packing at various velocities, pressure-taps were inserted below the gauze supporting the packing and above the top of the packing, and connected to a manometer. This arrangement gave very erratic results, probably because air bubbles collected under the gauze and much increased its resistance. Since the greatest conceivable Pitot effect was estimated to be negligible, it was decided to insert tubes directly into the packing. A "pressure-probe" of the type shown in Fig. VIII was used, the ends of the tubes being turned upwards to prevent any bubbles entering or blocking them. Although results were inclined to be erratic, they were very much more reproducible than with the original arrangement.

Calculation of Diffusion Coefficients. Several sets of samples having been taken with the flow velocity constant, a plot was made of

$$\log_{10} \frac{C_x}{C^*} \quad \text{vs.} \quad \frac{Y^2}{X}$$

$(\frac{cx}{c*})$ being averaged for all samples with the same v and χ) and the best straight line consistent with the "probability" equation was drawn through the points. Either the slope or intercept of the line gave the value of FD. Details of the calculation are given in Appendix F.

IV RESULTS.

IV RESULTS

The following is a table of the results obtained in this investigation

Packing type and size	Free volume	Superficial Velocity CMS/sec	Reynolds Number	Friction Factor	Pressure gradient Dynes/sq. CM/CM	Diffusion Coefficient CMS ^{2}/sec.
6 mm glass beads	0.36	0.25	11.99	61	0.0129	0.0103
		0.314	15.05	52.0	0.0174	0.0165
		0.32	15.31	51.5	0.0179	0.0200
		1.07	51.3	24.5	0.0956	0.0834
		1.66	79.5	19.5	0.183	0.196
		1.85	88.7	17.5	0.204	0.225
		2.30	110.	15.5	0.279	0.389
4 mm glass beads	0.36	0.28	8.93	75.5	0.0301	0.0105
		0.69	21.9	41.0	0.0995	0.0334
		1.22	38.9	29.0	0.220	0.0812
		1.75	55.8	23.3	0.363	0.133
		2.26	72.0	20.5	0.534	0.172
		2.36	75.3	20.0	0.568	0.224
*3/8" catalyst spheres	0.40	1.06	92.2	11.4	0.0240	0.0201
		1.06	92.2	11.4	0.0240	0.228
¼" Berl Saddles	0.59	1.91	97 (nominal)	32.8* see appendix B	0.0766	0.354

* 3/8-inch was manufacturer's nominal diameter. Displacement measurement of volume gave mean diameter as 11 mm.

V DISCUSSION OF RESULTS .

V. Discussion of Results

Accuracy of Experimental Work. The chief source of inaccuracy was the fact that the concentration distribution was not continuous; particularly at low rates of flow, the concentration varied sharply from point to point. Since the samples were withdrawn in effect from a point source, it was necessary to take a considerable number of individual samples and average their concentrations in order to achieve reliable results. The situation is analogous to that of plotting the density of shots on a target; if samples consist of small, constant areas of the target, the deviation between individual samples taken in the same region will be least at the centre, and will become larger as one goes outwards to regions of lower density. In the same way, the variation between individual samples taken in corresponding positions became greater as one moved further from the axis of the tower.

An experiment was performed to show the point-to-point variations in concentration. The five sampling tubes were arranged so that they formed two compact groups, the members of each group being very close together. The following results were obtained:

r	(cms):	0.97	1.16	1.40	....	2.87	3.10
$\frac{c}{c^*}$	(a) :	10.3	18.4	8.3	....	0.33	0
	(b) :	9.0	25.3	14.2	....	0.37	0

The samples (b) were taken immediately after (a), without disturbing the apparatus. It can be seen that the relative concentrations do not decrease uniformly as r increases, that large variations occur within short distances, and that the configuration changes somewhat with time.

In the above experiment (Runs 67 and 68) the value of x was 20 cms. and of the Reynolds' number 16.

It is felt that the error in the measurements of concentration was negligible, compared to those arising from the above effect, since they were reproducible to within about 3%.

A possibility of error lay in the fact that the linear rate at which samples were withdrawn through the sampling tubes was greater than the linear velocity through the tower (see Section III). The effect of this was to draw in liquid from regions remote from the actual sampling-point. Here, again, however, it is felt that since the velocity of withdrawal was at most 30 times the rate of flow, and hence the effective sampling area was only about 5 times that of the tube in diameter, the error introduced was small compared to that due to fluctuations in the concentration. Indeed, by averaging the concentration over a wider area than the cross-section of the sampling-tube, the sample-to-sample variation may have been reduced.

Other sources of error which have been considered, and which are felt to be probably unimportant are: the excess of the speed of injection of indicator over the flow velocity, and the influence of air bubbles on the effective shape and free volume of the packing (see section III.)

The nett error in the diffusion coefficient is difficult to estimate. From the consistency of the graphical correlation plots, it is considered that the maximum error is about 30%, and the average error between 5% and 10%.

The error in the measurement of the pressure-drop through the packing was of the order of ± 0·2 cms. of water for 20 cms. of packing. The reason for this error (when using the "probe" described in Appendix E) is not known; it obviously amount to a considerable relative error at low velocities and in the case of packings of low specific resistance, such as 3/8-inch spheres and Berl saddles. In future it might be desirable to use a considerably greater depth of packing to measure pressure-gradients.

Agreement with distribution equations. An inspection of graphs 9 - 18 will show that considering the discontinuities in the system, the distribution of concentration as found by averaging several measurements agreed remarkably well with either the Wilson or the probability equation. From an engineering point of view it is immaterial which law (if either) is followed exactly; however, the probability expression is more convenient, since it is easily integrated over a circular section; it is thus possible to calculate, for instance, the length of a tower of given diameter which is required, under given conditions, to "iron-out" or homogenise a fluid stream of non-uniform concentration (such a calculation is made, for example, on pages 21, 22 of Vol. I of the original lab. notes). Under the conditions obtaining in a catalytic reactor it is thought that abrupt discontinuities in concentration are unlikely to be encountered. In the first place, the Reynolds' number is likely to be such that considerable turbulent mixing will take place; secondly, the gaseous molecular diffusion co-efficient may be of the same order of magnitude as the coefficient of mixing due to flow, so that interdiffusion will occur between neighboring regions at all rates of flow.

Correlation of Results. Assuming that the magnitude of D depends only on the superficial velocity v the fractional free volume, F, the density ρ and viscosity μ of the fluid, and the diameter of the packing particles d, and that the tower diameter does not influence it (see

Appendix B), dimensional analysis shows:

$$\frac{D}{dv} \text{ is a function of } \left(F, \frac{dv\rho}{\mu}\right)$$

$$\text{or } \frac{D\rho}{\mu} \text{ is a function of } \left(F, \frac{dv\rho}{\mu}\right)$$

For the spherical packings, all but D, v and d are constant. Graph V shows a plot of D against $\frac{dv\rho}{\mu}$.

Since the friction factor f, defined for spherical particles as

$$f = \frac{\Delta P/L \cdot d}{2\rho v^2}$$

(where Δ p is the pressure drop in absolute units through a depth L of packing) is a unique function of the Reynolds' number (see Appendix B), D was also plotted against f (graph IV). Within the range of conditions covered by the experiments, these plots show that, approximately:-

$$\frac{D\rho}{\mu} = 0.035(Re)^{1.4}$$

$$\text{or } \frac{D\rho}{\mu} = 3(f)^{-2.36}$$

(F was constant at 0.36 for all the experiments except those involving 11 mm.beads and Berl saddles; hence its part in the correlation cannot be determined.) It is clear that whatever the exact form of these two functions, d can be eliminated between them; this would be true even for non-spherical particles, using arbitrary values of d. Thus for any given shape of particle, it must be possible to express D as a function of $\rho, \mu, v, \frac{\Delta P}{L}$ and of some dimensionless factor to account for variations in the fractional free volume. If it is desired to correlate data in which the shape of the particles and the fractional free volume vary independently of each other and of the other variables, then an empirical factor may have to be introduced to account for each of these. However, it appeared to the authors that shape and free volume might not appear in the correlation expression as independent variables, since their effects might be absorbed into $\Delta P/L$. Hence dimensional analysis was applied on the assumption that $\mathcal{D}$ is a function of $\Delta P/L$, ρ , μ & v only, and it was found that

$$\frac{\mathcal{D}\cdot\Delta P}{\rho\cdot v^3 L} \quad \text{is a function of} \quad \frac{\Delta P}{L}\cdot\frac{\mu}{v^3\,\rho^2}$$

Since μ and ρ were constant throughout, D was plotted against $\frac{\Delta P}{L\,v^3}$ (Graph VII)

For the spherical particles,

$$\mathcal{D} = 0{\cdot}42\left(\frac{\rho L v^3}{\Delta P}\right)\left(\frac{L\,v^3\,\rho^2}{\Delta P\cdot\mu}\right)^{-0{\cdot}2}$$

and the single value of $\mathcal{D}$ for Berl saddles was within 15% of that given by this expression. Although not conclusive, this is a promising result, since Berl saddles are far removed from the sphere in shape, and their fractional free volume was 1.6 times that for the spherical packings.

If further experimental work on non-spherical packings confirms the validity of this relationship, it should be extremely useful, since it would enable values of $\mathcal{D}$ to be predicted for such packings from pressure-drop measurements alone, and these are relatively easy to carry out or (in some cases) to find in the literature, while any expression involving the friction factor or Reynolds number suffers from the indeterminacy of these numbers for irregular shapes.

It is interesting to note that if the Reynold's number for 1/4-inch Berl saddles is calculated using this nominal diameter as d, the single value of D obtained for this packing falls close to the same line as that relating D to Re for spheres. However, it is not felt that this is more than a coincidence in view of the considerations put forward in Appendix B. In particular, the free volume for the saddles is much larger than for the spheres (0.59 and 0.36 respectively), and since there is every reason to believe that D depends on the free volume, it should enter into the correlation. In addition, there is some reason for thinking that the "nominal" Reynolds' number for Berl saddles does not correspond numerically with the Reynolds' number for spheres— i.e. that the change from turbulent to streamline flow does not occur over the same numerical range in the two cases. (See Appendix B).

Interpretation of Results. It does not appear feasible to provide a detailed and quantitative picture of the phenomena which lead to the type of mixing observed. There is good reason to believe that in all cases the flow through the packing was turbulent in some spots and streamline in others; hence one would not expect the diffusion co-efficients to bear any more simple a relationship to the Reynolds number in this region than does the friction-factor. It is suspected that at least two mechanisms are effective in this range— turbulent mixing or eddy-diffusivity, and "displacement" mixing; since the relative effects of the two mechanisms are probably varying in importance, a quantittative analysis would be difficult, even were it possible to predict the effect of one mechanism alone, which it is not. The fact that the simplified "geometrical" displacement co-efficient derived in Appendix B gives an approximation to the observed results at low rates of flow may or may not be significant. (Graph VI shows a plot of the values of D predicted by this expression for varying values of the Reynolds number; taking $F = 0{\cdot}36$, $\rho = 1$, $\mu = 0{\cdot}0125$ poises, the relationship has the form D = 0,00065 (R_e)). Further experiments at still lower rates of flow would be required to show whether the apparent agreement is coincidental. As far as could be observed in qualitative experiments, channelling, caused by fluctuations in the specific resistance to flow from point to point in the packing, is responsible for some of the mixing which took place. No quantitative theory of this effect is offered; it would be expected to lead to a "random-walk" type of distribution, but the value of the coefficient is difficult to estimate. It would not appear necessarily to lead to a coefficient of the same magnitude as that calculated for the "geometrical" coefficient.

The experimental results indicate that there is probably a mixing-effect even when the flow-velocity is so low (i.e. Reynolds' number less than 5) that flow is completely streamline. In such conditions point-concentrations would be expected to vary abruptly between zero and the original indicator concentration; the "mixing" would consist in the scattering of filaments of the indicator across the tower. In a regularly-arranged bed of isometric particles channelling could presumably not occur. It would be interesting to investigate such a system at low Reynolds numbers.

Comparison with Heat-Transfer Data. Since experiments on the transfer of heat in a packed tower through which a fluid is flowing are being carried out at the Institute at the moment, some remarks may be made on the bearing of the present work on the subject. The overall heat-transfer coefficient or apparent conductivity in a stream of gas passing through a packed tower is presumably due to three different types of mechanism —transfer due to the mass-transfer caused by the mechanism investigated in the present work;

nprmal thermal conductivity of the gas; and transfer of heat from gas to solid, conduction through the solid, and re-transfer to the gas beyond. The following expression includes a term for each mechanism:

$$K' = S\rho D + Fk_G + \phi(K_s, F, v, \rho, \mu, p)$$

where k' is the overall apparent conductivity, D is the apparent diffusion coefficient as measured in the present work, ρ the density of the gas, and S its specific heat. F is the fractional free volume - also equal to the fraction of any plane free from solids and hence available for gaseous conductivity; $\phi(K_s, etc)$ is a function of the conductivity of the material of which the packing is composed and of the velocity and physical properties of the gas.

Stadig (21) has shown that for air at a Reynolds' number in the neighbourhood of 100, the value of k' is of the order of 0.2 B.t.u / hr.-ft.- F., when the packing used was $\frac{1}{4}$-in. Berl saddles. Using our value of D for Berl saddles at a comparable Reynolds' number, it is found that the first term is about 1/10 of k', while Fk_g is about 1/40 of k'. Hence most of the transfer apparently takes place by the third mechanism - i.e. by the path gas-solid-gas.

VI CONCLUSIONS.

VI CONCLUSIONS.

(a) Mixing, in the sense of scattering of elements of fluid through the packing, occurs over the entire range studied - Reynolds' numbers from 10 to 110.

(b) The general form of the concentration-distributin in the tower at steady state was consistent with either the Wilson diffusion equation or with the probability equation derived from the "random walk" theory. These lead to slightly different expressions, but under the conditions of the experiments the difference between the two was less than the experimental error.

(c) The apparent diffusion-coefficient so determined increases with flow-velocity and with the diameter of the particles. For spheres:

$$\frac{D\rho}{\mu} = 0{\cdot}035(Re)^{1{\cdot}4}$$

over the range studied.

(d) The expression:

$$D = 0{\cdot}042\left(\frac{\rho L v^3}{\Delta P}\right)\left(\frac{\mu \Delta P}{\rho^2 L v^3}\right)^{0{\cdot}2}$$

which does not refer to particle diameter, correlates the single value obtained for Berl saddles reasonably well with those for spheres.

(e) The nature of the flow through the packing is considered to be viscous in some parts of the packing and turbulent in others over this range of Reynolds' numbers. Mixing is thought to be caused partly by local turbulence; partly by the divergences of flow caused by channeling (which is in turn caused by local fluctuations in the specific resistance to flow); and also, perhaps, partly by divergences caused by when the stream divides to flow past packing-particles.

(f) The distribution of concentration of the indicator in these experiments appeared to be discontinuous, particularly at the lower rates of flow, when it varied sharply from point to point; at higher rates turbulence seemed partially to even out these discontinuities. In all cases it was necessary to make and average many individual point measurements to obtain reliable results. There appeared to be quite a marked change in the appearance of the stream leaving the packing for relatively small changes in Reynolds' number, when the latter was about 40.

(g) As long as the diameter of the tower is more than five or six times that of the particles, it is concluded that none of the characteristics of flow (e.g., pressure-gradient, apparent diffusion-coefficient) are affected by the ratio of the two.

VII. RECOMENDATIONS FOR FUTURE WORK.

VII Recommendations for Future Work

Recommendations for future work will be made under the following heads --

(i) Variation of fluid properties and velocity.

(ii) Variation of packing.

(iii) Pressure-gradient measurements.

(iv) Elimination of air-bubbles from water.

(v) Qualitative experiments bearing on mechanism.

(vi) Improved methods of sampling.

(vii) Other modifications to apparatus.

(i) All the present work was carried out in the transition region between streamline and turbulent flow. It would be of interest to extend the measurements to the purely streamline and purely turbulent regions. The former requires an improved sampling technique, the latter various changes in the apparatus. The viscosity of the water might be altered considerably by adding small quantities of cellulose ethers. Finally the investigation should be extended to gases.

(ii) Further work on spheres of different sizes, on Berl saddles and on irregular particles is required, particularly to see whether the mixing can be correlated with the pressure gradient. It would be of interest to investigate the effect of changes in free volume in the case of a packing (such as Berl saddles) where this can be varied independently of other factors.

iii) Correlation with pressure gradient requires an improved technique for measuring the latter, with greater accuracy than the method used by the present authors. It may well be advisable to measure pressure-drops through considerably greater depths of packing.

(iv) The formation of air bubbles on the packing may prove to be serious in some circumstances-e.g. when using a very fine packing. The trouble could be eliminated, it is thought, by giving the water from the taps no chance to warm up while on its way to, or in passage through, the tower. Thus the constant head tank and all pipes might be lagged, and the tower itself waterjacketed. The addition of a wetting agent to the water might prevent bubbles clinging to surfaces in the tower.

(v) The use of a two-dimensional transparent model might give some insight into the mechanism of the mixing process -- e.g. cylindrical blocks between two sheets of glass, or standing upright in a shallow tray through which water flows. If cryolite crystals, which have the same refractive index as water, were used as the packing particles, it should be possible to see a good deal more of what is going on than when glass beads are used.

(vi) As has been mentioned, point sampling is unsatisfactory, particularly at low Reynolds numbers, because of the point to point fluctuations in concentration. This means that a large number of samples must be taken and their concentrations averaged. It is suggested that <u>area</u> sampling be used. It has been shown that a standard probability distribution is followed; the integration of this expression over a circular area concentric with the injection-axis is simple. All that is required is a method of finding the mean concentration of indicator in the fluid flowing through a given circular cross-section. A suggested experimental arrangement is shown in Fig. XI. The diameter of the sampling-tube should be carefully chosen with regard to conditions so that approximately half the indicator flows through it, otherwise results will not be accurate.

(vii) If packing-particles of larger diameter than those used by the present authors are used, it would be advisable to construct a tower of greater diameter. To obtain reasonable results samples must be taken a good many packing-diameters from the injection point; on the other hand, samples must be taken at cross-sections where the concentration at the wall is small compared to that at the centre. These two requirements will be incompatible if the packing diameter is more than about 1/10 that of the tower. It may also be advisable, particularly when using packings of low specific resistance, to use a larger calming section at the bottom of the tower to ensure that flow is symmetrical. The container used for the purpose was felt to be too small to act efficiently at high rates of flow.

VIII APPENDICES.

APPENDIX A

The Geometry of Spherical Packings and Derivation of a Geometrical mixing Coefficient.

If a mass of spheres of uniform size is placed in a glass vessel, it can be seen that the arrangement of the spheres is not regular. Tapping the vessel or running a rod through it increases the regularity of the packing on the lower parts of the wall and decreases the volume of the aggregate; however, it can be seen that the arrangement of the spheres in contact with the upper parts of the walls, and of the spheres at the surface, is far from regular. It appears that the system as a whole departs widely from any geometrically regular system of packing; experiments designed to find the free volumeF indicated that the closeness of packing varies considerably with variations in the way in which the system is treated (e.g., tapped, stirred, rammed).

The sphere-diameter, d, and the fraction of the total volume not occupied by the substance of the spheres, F, are the important numerical characteriestics of a spherical packing. d, of course, is quite definite and constant. F, which may be expected to have an important influence on fluid flow through the packing is, as already remarked, dependent on various factors.

It is felt however that under the conditions of the experiment its value was essentially constant, since the packing was thoroughly stirred in situ under water between each run, so that the spheres may be assumed to have settled into their equilibrium position under much the same conditions each time.

Neglecting for the moment the effect of the wall of the confining vessel, we may regard the packing to be isotropic and uniform, in the mass. If the fraction of free volume is F, and the number of spheres in unit volume is N, we have

$$F = 1 - \frac{1}{6} N\pi d^3$$

Consider a plane drawn through the packing; the fraction of the area of this plane lying outside the spheres is A. Hence in a lamina of thickness dh, the free volume will be Adh. Thus in a finite volume composed of a very large number of such laminae, in each of which the free volume is Adh, the total free volume will be $\int_0^h Adh = Ah$. Since the total volume will be h, we have

$$F = \frac{Ah}{h} = A$$

i.e., the fractional free area in any plane is equal numerically to the fractional free volume of the mass.

By a similar argument it can be shown that if a straight line is drawn through the packing, the fraction of its length which lies outside the spheres is equal to F. Such a straight line will cut every sphere whose centre lies in a cylinder centered on the line and of diameter d (the sphere diameter). The volume of such a cylinder will be $\frac{\pi d^2}{4}$ units per unit length, and since there are N spheres per unit volume, and hence N sphere-centres per unit volume, the line will cut $\frac{N\pi d^2}{4}$ or $\frac{3(1 - F)}{2d}$ spheres per unit length.

Similarly, unit area of any plane cuts all the spheres lying in a volume of 2d, which is $\frac{2(1 - F)}{\pi d^2}$ spheres.

Furnas (1) argues that the effective free area for the flow of a fluid is not F. He concedes that the fractional free area of a plane drawn through the packing would be F, but argues that this is not the cross-section available for fluid flow, since " 'free area' is by no means all effective in allowing the passage of (fluid) as the bulk of the area is blocked by overlying pieces of solid material". He mentions, but does not reproduce, geometrical considerations which show the effective free area for spherical packings to be a fraction of F. The authors are unable to agree with his argument, so far as they understand it, and consider that at any rate for smooth, non-re-entrant shapes such as spheres the free area must be taken as F.

Furnas devotes a good deal of attention to the effect of the wall of the confining vessel on the pressure-gradient--flow-rate relationship of the packing. There is a prima facie case for supposing that the effect of the wall is to increase the free volume in the layer of spheres immediately adjacent to it, and thus to offer a path of lower resistance than through the main body of the packing. Consider a layer of spheres resting on a plane; there is a considerable amount of free volume between the plane and the spheres. If now the surface of the plane were embossed with hemispherical humps, these would partially fill the spaces between the spheres and the plane, and reduce the free volume. This latter arrangement may be supposed to approximate more closely to the conditions in the body of the packing. However, as has already been mentioned, the spheres in contact with the walls tend to be more regularly arranged, as far as can be seen, than those in the body of the packing; under the conditions of our experiments, quite a close approach to hexagonal packing was attained over the major part of the tower-wall. This would tend to reduce the free volume in the outer layer. A calculation has been made to find the free volume in a layer of thickness d/2 adjacent to a plane on which spheres of diameter d are lying in hexagonal (i.e., closest possible) packing (see Fig.I); The value of F in this layer is about 0.4, and since the value of F in the body of the packing is about 0.36, the difference is not great. More will be said in Appendix B on this subject in relation to flow. A number of experiments were carried out to find the overall free volume in packing confined in vessels of various shapes and sizes in which the ratio $\frac{\text{wall area}}{\text{total volume}}$ varied widely, with a view to extrapolating to zero value of this ratio to find the value of F in the body of the packing. The earlier experiments appeared to show a trend of the type which would be expected--i.e., F decreased as the proportion of wall area to volume of the vessel decreased. At this time, however, the effect of the treatment of the packing on F was not fully realized. A later set of experiments, conducted in cylindrical vessels, in which the packing was consolidated under water to a constant volume by a standardized method of ramming before F was determined, showed that the value of F was substantially constant at 0.36 - 0.37 provided that the diameter of the containing vessel was more than say five times greater than the diameter of the packing.

It is considered that the increase of free volume above a certain value of the diameter ratio is

Table

Free Volumes for Glass Spheres in Cylindrical Towers.

$\frac{\text{Particle diameter}}{\text{Tower diameter}}$	F
.247	0.42
.225	0.43
.150	0.37
.130	0.36
.064	0.36
.043	0.36

due to an effect on the mode of packing in the interior of the vessel, rather than to an excess of voids in the immediate vicinity of the wall.

The 11 mm. spheres (nominal diameter 3/8 ins.) gave a value of F of 0.4; ratio of particle to tower diameter was 0·12. But the particles were not entirely uniform in size and shape, which may account for this high value.

The free volume was determined in one of two ways:

(a) Some water was poured into a cylindrical vessel, some packing added and consolidated, and the apparent volume, V_1, of the packed region and total volume V_2, of packing and water measured. More packing was then added and consolidated as described; the apparent (V_3) and total (V_4) volumes were noted again. Then —:

$$F = 1 - \frac{V_4 - V_2}{V_3 - V_1}$$

(b) Packing was consolidated under water in situ in the tower of the apparatus. Water was run off from a tap at the bottom of the tower until the surface stood at a certain height in the tower; a measured volume of water was then run off,and the fall in height of the water surface was observed.

Both these methods eliminate the "end-effects" which might be expected at the bottom and top of the vessel.

Derivation of a "Geometrical" Mixing Co-efficient.

Preliminary consideration of the mixing of fluids in a bed of spheres led to the idea of basing a derivation of a "mixing-coefficient" on premises which, while probably unrealistic, might yet give an idea of the order of magnitude of the actual co-efficient under limiting conditions. The derivation is based on the idea of the splitting of streams in streamline motion through the bed--discussed in the Introduction and illustrated in Figure IV (ii).

Consider an inelastic particle of negligible size falling through a bed of spheres whose diameter is d. The particle will move in a vertical straight line until it strikes a sphere; it will then "skirt" the sphere in a great circle path (taking the shortest route to the edge) until it rolls off and falls in a straight line to the next sphere.

It has been shown earlier in this appendix that a straight line drawn through a bed of spheres cuts $\frac{3}{2} \cdot \frac{(1 - F)}{d}$ spheres per unit length, where F is the fractional free volume. It was also shown that the fraction of the length lying outside the spheres is F. Hence the mean free path between spheres is $\frac{2Fd}{3(1 - F)}$.

A little consideration will show that the path of the particle considered can cut no new sphere while it is skirting a sphere, whereas a straight line drawn through the packing can cut no new sphere while it is "inside" a sphere. Hence

$$\frac{\text{Free length of straight line}}{\text{total length}} = \frac{\text{total length} - \text{length inside spheres}}{\text{total length}}$$

$$= 1 - \frac{\text{length inside spheres}}{\text{total length}}$$

$$\frac{\text{Free length of particle path}}{\text{Total vertical dist. dropped}} = \frac{\text{Total vertical dist.} - \text{vertical dist. dropped in skirting path}}{\text{Total vertical distance}}$$

$$= 1 - \frac{\text{Vertical distance dropped in skirting path}}{\text{Total vertical distance}}$$

As Figure II is intended to convey, if a vertical straight line strikes a sphere at a certain spot, the length of the line lying within the sphere will be twice the vertical distance dropped by the particle, if it strikes the same spot, while it is skirting the sphere. Since a straight line cuts $\frac{3}{2} . \frac{(1 - F)}{d}$ spheres per unit length, and (1 - F) of of its length lies inside the spheres, the mean length of its path through a sphere is $\frac{2d}{3}$. Hence the mean vertical distance dropped by the particle while skirting a sphere is half of this, or $\frac{d}{3}$. Since the number of spheres struck by the particle per unit length of *free* vertical path is the same as the number cut by the straight line per unit length of *free* path, it can easily be shown that the number of spheres struck by the particle in unit length of total vertical path is $\frac{3(1 - F)}{d(1 + F)}$.

The projection of a sphere onto a horizontal plane is a circle of diameter d, and the particle may fall anywhere within this circle. The probability that it falls at a distance r from the centre of the circle is $\frac{8\pi r.dr}{\pi r^2}$ or $\frac{8r.dr}{d^2}$, hence the average value of the distances from the centre for all possible points of impact is $\int_0^{d/2} r \frac{8rdr}{d^2} = \frac{8}{d^2}\int_0^{d/2} r^2 dr = \frac{d}{3}$

Hence $\frac{d}{6}$ is the mean horizontal displacement undergone by the particle per impact. Now suppose that the particle moves at a constant vertical velocity u. In unit time it will undergo $\frac{3w(1-F)}{d(1+F)}$ horizontal displacements of mean value $\frac{d}{3}$. These displacements will be completely random in direction, in that the direction of any one will be unrelated to that preceding it. Under these circumstances we can apply the "random walk" theory to calculate the mean net horizontal displacement of the particle in a given time. (The theory is discussed in Kennard's "Kinetic Theory of Gases".)( )The expression for displacements occurring in three dimensions (as in a gas) is

$$\frac{\overline{x}^2}{t} = 2\left(\frac{\ell}{\sqrt{3}}\right)^2 Z$$

where $\overline{x}^2$ is the mean square of the net displacement, ℓ is the mean length of one displacement, t the time, and Z the frequency of displacements. In a gas, an individual displacement may be in any direction in space, and $\frac{\ell}{\sqrt{3}}$ is the mean of the projections of the individual displacements onto a single straight line. The displacements which we are considering take place horizontally only, so that the mean of the projections of individual displacements onto a given line will be $\frac{\ell}{\sqrt{2}}$, ℓ having the value already calculated, $\frac{d}{6}$. Hence for the particle under consideration,

$$\frac{\overline{x}^2}{t} = 2\left(\frac{d}{6\sqrt{2}}\right)^2 Z$$

Since Z had been shown to be $\frac{3u(1-F)}{d(1+F)}$,

it follows that $\frac{\overline{x}^2}{t} = \frac{du(1-F)}{12(1+F)}$

If a large number of particles move through the bed in the way described, the result will be that a process analogous to diffusion will take place horizontally. It can be shown (see Kennard's "Kinetic Theory of Gases")(22) that the diffusion co-efficient D will be

$$D = \frac{\overline{x}^2}{2t}$$

hence for the system considered

$$D = \frac{du(1-F)}{24(1+F)}$$

for horizontal diffusion.

For particles of other than spherical shape, a similar (but more difficult) treatment would also give an expression of the form

$$D = K \times du$$

where K is a constant.

The free volume of the 4-mm. and 6-mm. spheres used in this work, F= 0.37. Substituting in the expression given above $u = Fv$ and $(Re) = dv\rho/\mu$, we have

$$D = \frac{dv}{F} \cdot \frac{1}{24} \frac{(1-F)}{1+F} = \frac{1}{F} \cdot \frac{1}{24} \cdot \left(\frac{1-F}{1+F}\right) \cdot \frac{\mu}{\rho} (Re)$$

Substituting F= 0.37, we have D = 0.00065 (Re). This expression is plotted on Graph VI, where it is compared with the line obtained by extrapolating the experimental plot of D vs. (Re) .

APPENDIX B

Fluid Flow through Packed Towers.

The treatment of fluid flow through beds of solids is much more complicated than for that through geometrically simple conduits, and the correlation of experimental data has been at the best semi-empirical, with the theoretical background of the expressions open to some doubt. Three papers by Furnas (1), Chilton and Colburn (2) and Meyer and Work (3) present most of what is known on the subject. A summary is given in Perry's "Chemical Engineer's Handbook", (9).

The basis of the correlation equations is a modified Reynold's Number (hereafter referred to simply as the Reynold's number):

$$(Re) = \frac{d v \rho}{\mu}$$

and the Fanning Equation:

$$\Delta P/L = \frac{2 f v^2 \rho}{d}.$$

where d = particle diameter (actual or fictitious), v = superficial fluid velocity--i.e., linear rate of flow if packing were removed from containing tower.

Consider a fluid flowing through a bed of spheres at a velocity so low that turbulence is ruled out; and follow a single particle in the fluid through the bed. At any instant it can be pictured as flowing through a conduit of irregular and changing cross-section.

If it is near the center of such a conduit it will be travelling faster than if it is near the wall; a section through the bed would show a number of such "conduits", where flow is brisk, surrounded by zones of decreasing velocity, with stagnancy at the surfaces of the spheres. However, the maximum velocity through all these conduits is not equal; fluctuations in the arrangement of the spheres provide "channels", which are conduits whose cross-sections are by chance larger than the average for a considerable distance. These channels are not continuous throughout the depth of the bed (unless flow-rates are high enough to lift the spheres), but peter out and are replaced by others in other parts of the cross-section. In any cross-section, therefore, there are regions in which the maximum velocity is much higher than the average, and others in which it is much less. As the location of these channels varies from section to section, the particle will wander with respect to its original axis of flow as it passes through the packing. Moreover, two particles which are originally close together may become widely separated in this fashion. This type of behavior is in marked contrast to that of a fluid flowing past an isolated sphere in streamline motion.

The flow-paths of particles of fluid in this latter case are shown in Figure IV (1); it can be seen that any particle has the same situation with respect to the axis of the sphere before and after the encounter, so that no displacement or splitting of the stream can occur. Mixing by channeling does not exclude mixing by the mechanism illustrated in Figure IV (ii) and discussed in the Introduction.— i.e "displacement" mixing.

As a further consequence of channeling, it may occur that a stream in a conduit reaches a relatively high velocity and then debouches into a region where the velocity is low. As a result, turbulence may be set up locally even when flow in most regions is streamline.

This picture of the nature of the flow through the packing is supported by some qualitative experiments carried out with 6-mm. glass spheres in an 8-cm. glass tube. A layer of the spheres 4 or 5 diameters thick was supported on a wire gauze, and water was run through the tube at such a velocity that flow in the open part of the tube was steamline. This was confirmed by injecting a solution of methylene blue through a capillary tube beneath the layer of packing; the linear rate of injection was less than that of flow, so that the dye was drawn out into a smooth filament. The following phenomena were observed:

(a) On several occasions the filament, after broadening to a ribbon a few millimetres wide, split into two distinct parts (with apparently no colour in the intervening region), which then diverged and entered the packing at points some 6-8 ball diameters apart, well off the axis of the tube.

(b) Filaments which entered the packing emerged from the top either distorted into broad and irregular sheets, or split into several streams; the edges were sharp, and remained so up the tube. The extreme distance apart of any two parts of the visible colour was not more than 3-4 sphere diameters.

(c) The coloured streams emerging from the top of the packing did not emerge in a direction parallel to the axis of the tube, but inclined at small and various angles to it. At steady state an emergent filament appeared as a stationary line of a curved or spiral shape, tending to straighten out higher up the tube.

(d) When the coloured filament first entered the packing, it could be seen to accelerate, and the emergent streams could be seen to be moving considerable faster than the average velocity of the water in the open tube. Under some conditions, turbulence could be seen in the shape of very fine, uniform ripples at the boundary between an emergent filament and the relatively stagnant fluid surrounding it.

Figure V shows a typical situation.

When the rate of flow was increased to the point at which the flow in the open tube was turbulent, the filament leaving the injector tube was considerably shredded and contorted before reaching the packing. Instead of passing steadily through one point in the bed, it oscillated rapidly over the whole surface. In the course of routine experiments it was observed that at higher rates of flow some kind of oscillation or fluttering of the coloured fluid was going on in the packing near the injection-point. There was a marked change in appearance of the fluid leaving the top of the packing as the velocity was increased. At low velocities the colour emerged in streaks or filaments -- i.e., there were discontinuities in the concentration; at high velocities the colour distribution from point to point was continuous. The change occurred at a Reynold's number (calculated for the packing) in the neighborhood of 40.

When using 11-mm. white alumina spheres, observations could be made on filaments of colour which channeling brought to the wall of the tube. It could be seen that providing Re was not too high, an almost steady state was reached, the position of a filament hardly changing, except for local turbulence in spots. The path of such a filament was very devious. It appeared to be governed by channeling effects, rather than by purely geometrical displacements, such as those considered in Appendix A.

Whatever the exact nature of the mechanism of flow through pacing, it is generally admitted that there is no abrupt transition from streamline to turbulent motion. For spheres the flow is entirely streamline below Reynolds Number = 5 (3); the pressure-gradient through the packing is then proportional to the velocity--i.e., the friction factor f in the Fanning equation is inversely proportional to the Reynolds Number. At sufficiently high Reynolds numbers (the lower limit is not accurately known), flow appears to become entirely turbulent, and f is proportional to the (-0.15) power of Reynolds Number. There is a transition region where the exponential relationship between f and Re varies continuously (see Graph I and Perry (9))

All the experiments conducted by us were probably in the transtion region;--values of Re (Reynolds Number) lay between 10 and 110.

It is suggested that the behaviour of the coloured filament immediately before and after entering the packing in the experiments described shows in an exaggerated form the processes occurring within the packing--splitting by channeling, and localised turbulence which may lead to eddy-mixing. It cannot be said that this picture of the flow gives any reason to suppose that the geometrical mixing co-efficient derived in Appendix A will correspond to the facts; however, experiments show this to be actually a fair approximation for the value of the co-efficient observed at low rates of flow (see Graph VI).

As regards the influence of the wall of the confining vessel on the distribution of the elements of the packing, and hence on fluid flow, it has already been stated in Appendix A that, for the case of spheres, both geometrical considerations and actual measurements of F indicate that the free volume in the part of the packing adjacent to the wall is equal or very nearly equal to that in the body of the packing. Furnas (1) derived a semi-empirical expression for a co-efficient A to be used in a modified form of the Fanning equation:

$$\frac{\Delta P}{L} = \frac{f' A_f \cdot v^2 \cdot \rho}{2d}$$

where f' is supposed to be a factor which is the same for all sizes of particle at a given Reynolds Number, while A_f is a function of the ratio of the particle diameter to the tube diameter, which corrects for the influence of the tube wall on the free volume of the packing (Perry (9)).

The authors' experiments on free volume, already referred to, indicated that the overall free volume is constant as long as the tube diameter is at least 5 times as great as the particle diameter. Moreover, both White (7) and Uchida and Fujita (8) found that the pressure-drop was uninfluenced by the ratio of particle diameter to tube diameter as long as this was less than 1/6.

The authors tried to perform a critical experiment on this question. The tower of the apparatus was packed normally with 6-mm. glass spheres, water was run through at an ascertained velocity, and the pressure-gradient through the packing observed. A thin sheet of metal was then rolled into a rough cylinder or spiral scroll, and thrust co-axially into the packing in the tower. The area of the metal surface (counting both sides) was about equal to the area of the tower wall in contact with the packing; if, therefore, the layer of packing immediately adjacent to a flat surface offers a path of lower specific resistance to the flow of liquid than the body of the packing, one would have expected the pressure-gradient to be lower, at the same velocity, than before. Unfortunately, owing to the uncertainty involved in the pressure-gradient measurements (see Appendix E), all that can be said is that if there was an effect it was about of the same order of magnitude as the experimental uncertainty. One would have expected, following Furnas(1), that the pressure through the packing would drop by some 0.1 - 0.2 inches of water, but unfortunately the measurements were not reproducible to a greater accuracy than this.

Chilton and Colburn (2) used the factor A_f given by Furnas in correlating their experimental results and those of previous workers. As far as their own data are concerned, we have re-plotted them without reference to A_f --i.e., plotting f, as calculated from the normal Fanning equation, against the Reynolds Number. It is found that for those experiments in which the ratio of particle diameter to tube diameter was less than 1/6, the correlation obtained was as good as, or better than, that obtained when using A_f ; when this ratio was greater than 1/5, the friction factor was markedly low.

Finally, the authors have measured pressure-gradients and calculated from them friction-factors (without using A_f) for 4-mm. and 6-mm. beads in a tower 9 cms. diameter. The friction-factors for given Reynolds' numbers for the two sizes are very close together (see Graph I); it is true that the values of f for the smaller size appear to be some 5% higher than for the larger size, but in view of the errors in the pressure-gradient measurements, it is doubtful whether this is significant.

To sum up, our own free-volume measurements, Chilton and Colburn's data, and the opinions of other investigators appear to lead to the conclusion that as long as the diameter of the confining tube is 5 or 6 times as great as the diameter of the packing particles, variations in the former will not affect the flow through the packing.

This implies that in these experiments the linear rate of flow was substantially constant, in the mean, in all parts of the cross-section of the tower. The matter has been stressed because it is of some importance in correlating any measurements concerned with flow through packings. In particular, the equations the authors used to calculate the diffusion co-efficients from concentration measurements were derived on the assumption that the mean linear rate of flow is uniform across the tower. Were this not so, a correction would have to be applied in order to calculate the true mean linear velocity from the volumetric rate of flow.

The treatment of non-spherical particles is more difficult, and it is necessary to define their diameter arbitrarily.

White (7) has correlated pressure-drop data for Raschig Rings and Berl saddles in the following way: for a given shape of packing, an empirical relationship is drawn up between the nominal diameter d_p of the particles and a corrected diameter d_p/f_A. The nominal diameter is used to calculate the Reynold's number, and then the friction factor corresponding to this is read off the Chilton-Colburn curve. The pressure-gradient can then be calculated by using the Fanning equation, with this value of the friction factor, and d_p/f_A in place of the diameter. In other words, the pressure-gradient for an irregular shape is equal to that for spheres at the same Re (using nominal diameter of particles), multiplied by the factor (f_A); or we may say that the friction factor for an irregular shape is that for spheres at the same Re, divided by (f_A) using the nominal particle diameter throughout. The data for Berl saddles are scanty, but for rings the procedure is justified in that the f vs. Re curve "breaks" at the same value of Re as for spheres. This may not be so for Berl saddles; White's data are all in the turbulent region and do not cover the "break". (See also the data on 1/4-inch Berl saddles, plotted on Graph II.) Formulae for f_A for rings and saddles are given in Perry (9).

It would be more satisfactory, when dealing with non-spherical particles, to use a correlating expression containing no reference to particle diameter, and an attempt has been made to derive one (see "Correlation of Results").

If the free volume F can vary independently of the particle shape, then F must also be introduced into the correlating expressions. The subject was studied by Meyer and Work (3); see also Perry (9). This problem, however, did not occur in this work.

Appendix C

Derivation of "Probability" Equation for computation of Diffusion Coefficient from Concentration Measurements. Comparison with Wilson Diffusion Equation

In the course of flow through a packing, mixing may be expected to to occur in several ways--e.g. molecular diffusion, eddy diffusion, and mixing caused by the geometrical characteristics of the packing. These are all alike in that they are caused by displacements of elements of the fluid which are large in number, random in direction, and small compared to the mean total displacements to be measured. The effectiveness of these various processes in causing mixing depends on the frequency and average length of these random displacements; in the case of a liquid, the mean free path of a molecule in thermal motion is so small that prolecular diffusion plays a negligible part under under the conditions of experiment.

The distribution of a number of particles which, starting from a Common Centre, have undergone displacements of the kind described (random, numerous, small) is given by the "normal probability function". This function expresses the probability of a particle which was originally at the origin being now in a plane which cuts the x - axis perpendicularly at x: ---

$$P_x = \frac{1}{\sqrt{2\pi \bar{x}^2}} e^{-x^2/2\bar{x}^2} \cdot dx$$

where $\bar{x}^2$ is the mean square of the x- coordinates of all the particles. Similarly:

$$P_y = \frac{1}{\sqrt{2\pi \bar{y}^2}} e^{-y^2/2\bar{y}^2} \cdot dy$$

for displacement along the y- coordinate. This if displacement along the x & y coordinates only is considered, & if we take $\bar{x}^2$ equal to $\bar{y}^2$ at any time (i.e. diffusion isotropic) we have

$$P_{x,y} = \frac{1}{2\pi \bar{x}^2} e^{-r^2/2\bar{x}^2} . dx . dy$$

where $r^2 = x^2 + y^2$

and P_{xy} is the Probability of the x & y coordinates having simultaneously the values (x, y).

Consider a very thin stream of an indicator fed into the axis of a stream of moving fluid, & suppose for the moment that diffusion takes place only in directions at right-angles to the direction of flow. Then the above expression gives the probability of a given molecule of the indicator being at a specified point at a distance r from the axis, when $\bar{x}^2$ refers to all the indicator molecules entering the stream at the same instant.

It can be shown (see Kennard(22)) that the co-efficient of diffusion, D, will be given by the expression

$$D = \frac{\bar{x}^2}{2t}$$

where t is the time which has elapsed since the indicator entered the stream. (The validity of this expression is discussed later in this appendix). Hence

$$P_r = \frac{1}{4\pi t D} e^{-r^2/4tD} . dx . dy$$

for the probability of a particle being at a specified point distant r from the axis, in a plane distant ut from the point of entry, where u = linear flow velocity (see fig. X)

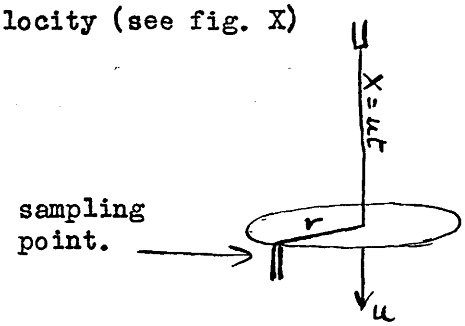


Fig. X

Since all the indicator molecules entering the stream at a given instant reach this plane at the same time, the expression for P_r also gives the fraction of the total number of indicator molecules entering the stream which pass through the differential area dx. dy. Thus if N molecules enter in unit time, then NP_r pass through this area in unit time. For the purposes of measurement, a sampling tube of small but finite cross-section a is used; it is assumed that P_r is effectively constant over this area.

In any plane drawn through the packing perpendicular to the direction of flow the free area unoccupied by the subtance of the packing is a fraction F of the total. The average linear velocity across the plane will then be v/F, where v = V/A, V being the volumetric rate of flow, & A the total area. Thus in unit time a volume av/F of fluid flows through an element a of free area. Now P_r expresses the probability of a particle being in a given overall area of the cross-section; since some of this area is occupied by the substance of the packing, & the particle cannot be in this part, the probability of it being in in an equal element of free area is P_r/F. Thus the total number of indicator molecules passing through a small element a of free area in unit time is $\frac{NaPr}{Fdx.dy}$, and their concentration is therefore

$$C = \frac{N P_r}{v . dx . dy.} = \frac{N}{4\pi t D v,} e^{-r^2/4tD}$$

If the sampling - point is situated in a plane distant X from the point at which the indicator enters,

$$X = ut = \frac{vt}{F}$$

also N = C * V, where C* is the average concentration of indicator in the entire fluid stream. Hence

$$\frac{C_x}{C^*} = \frac{V}{4\pi F D} e^{-r^2 v/4FDx}$$

It can be seen from the derivation of this expression that if sampling is continued at a constant rate for a finite time, & if C is the mean concentration of the sample so collected, while C* is the mean concentration of the effluent over the same period; then providing all the other factors remain constant, variations in the rate of injection of indicator are inmaterial.

This expression is only valid so long as diffusion in directions parallel to the axis of flow can be neglected without error. Otherwise, similar reasoning leads to the expression:

$$\frac{C}{C^*} = \int_{\xi=0}^{\xi=\infty} \frac{FV}{v^2}\left(\frac{v}{4\pi F D \xi}\right)^{3/2} e^{\frac{-\left[r^2+(x-\xi)^2\right]}{4FD\xi}} . d\xi$$

However, the simpler expression seems justified by the experimental results.

Previous workers (4, 5) have employed, the Wilson Equation for the evaluation of D. This equation was originally derived (6) for the conduction of heat from a point source through an isotropic stream of moving fluid, but owning to the mathematical identity between the fundamental equations for heat transfer & for diffusion, can be applied with equal validity to the latter. Like the probability equation, it neglects the effect of diffusion in the direction of flow.

For an open tube, the Wilson Equation has the form:

$$C = \frac{N}{4\pi D\sqrt{x^2+r^2}}\, e^{\frac{-u\left(\sqrt{x^2+r^2}\,-x\right)}{2D}}$$

Correcting for the space effect of the packing & transforming by multiplying top & bottom of the exponent by $\left(\sqrt{x^2+r^2}+x\right)$, we have

$$C = \frac{N}{4\pi FD\sqrt{x^2+r^2}}\, e^{-vr^2/2FD\left(\sqrt{x^2+r^2}+x\right)}$$

$$\text{or} \quad \frac{C\sqrt{x^2+r^2}}{C^*} = \frac{V}{4\pi FD}\, e^{-vr^2/2FD\left(\sqrt{x^2+r^2}+x\right)}$$

This form show that the expression is identical with the Probability Equation when $r = 0$, & that it will approximate it closely when $r^2 \ll x^2$ Since under our experimental conditions r^2 was never more than 1/10 of x^2 (& usually much less) $\sqrt{x^2+r^2}$ was never more than $x\sqrt{1.1}$, or 1.05 times x. Hence the maximum variation in the co-efficients would be 5%, in the exponents 2.5%, as between the two expressions. The experimental error was greater than this, so that no comparison can be made between the two.

The Probability Equation was used because its derivation was more fully understood, because it appears to correspond to the mechanism of the mixing process, & because it is mathematically somewhat simpler than the Wilson Equation (e.g. it can be easily integrated). Assuming that both the Wilson Equation & the Probability Equation have been derived from their starting assumptions without error, the discrepancy can only be due to the non-validity of the expression:

$$D = \frac{\bar{x}^2}{2t}$$

where $\bar{x}^2$ is the mean square displacement undergone by an element of fluid in time t . It was by using this expression to substitute for $\bar{x}^2$ in the "random-walk" distribution that D was introduced into the probability equation. The derivation of this expression is given in Kennard (22). In the course of the derivation it is tacitly assumed that the concentration gradient in any direction is constant; since, in fact, in the case under consideration the gradient varies in all three dimensions, a discrepancy will arise from this assumption, and D in the diffusion equation is not, in fact, equivalent to $\bar{x}^2/2t$ in the random walk theory in such a case.

Since the probability equation has been used throughout, D is defined as $\bar{x}^2/2t$; however, a numerically equal value would have been obtained had the Wilson equation been used, and D defined accordingly. More accurate experiments would be required to determine which expression more nearly corresponds to the facts; it is clear that neither can be more than an approximation, since both lead to finite values of the concentration at all distances from the axis, while practically the concentration falls to zero at a finite distance. Moreover, both are continuous functions, while actual diffusion of any kind involves the movement of discrete elements of matter. A general survey of the validity of the diffusion and proability treatments is given in the following notes.

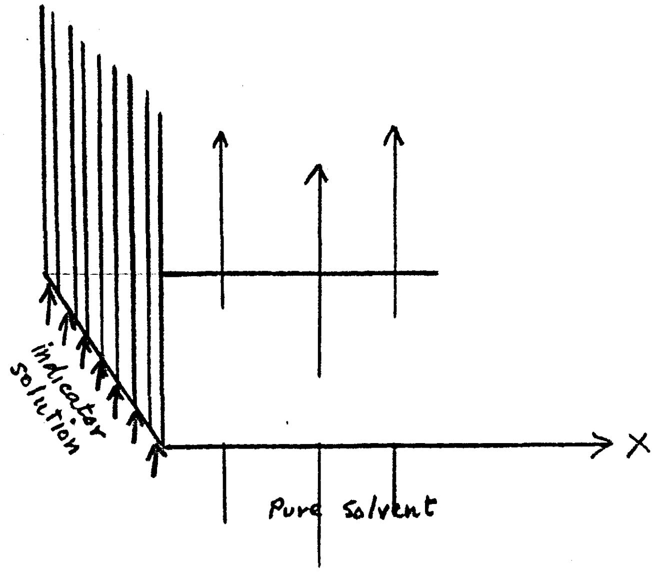
(a) The diffusion equation is an arbitrary or empirical expression with no theoretical basis. The expression which it gives for the rate of transfer at a point refers only to the concentration-gradients at that point, and not to conditions in other parts of the system.

(b) The probability equation is derived from the random walk theory, and will be strictly true when transfer is due to random displacements of the type specified in that theory.

(c) For the special case when the concentration-gradient on any axis in the system is constant along that axis (extrapolated to infinity), it has been shown by Einstein thatthe probability and diffusion-equations are identical in form, and that D in the former is then equivalent to $x^2/2t$ in the latter.

(d) However, for the general case the two are not equivalent; it is clear that they cannot be, since the diffusion equation refers only to conditions at a point, while to express in terms of the random walk theory the quantity of material crossing a given plane requires a knowledge of the concentration of the substance at all distances away from the plane in both directions. (The fact that in the Wilson equation, for instance, the difference is not great is because in fact it is those parts of the system which are close to the plane in question which contribute most to the transfer across it; the error in assuming the concentration-gradient constant over a short distance will not be great, while at greater distances the contribution to the total will be so small that large errors in assessing this contribution are of little importance).

(e) Thus the diffusion equation is only strictly applicable in cases where the physical conditions are such that the concentration-gradient is constant along any given axis. It is clear that this can never be the case when a solution containing a finite concentration of the diffusing substance is injected into the pure solvent.



If the arrangement used in the present work were replaced, for instance, by that shown above, indicator solution being injected from a linear source, and the direction of diffusion being limited to the x-axis, it is clear that the concentration-gradient on the latter would vary with x .The diffusion and probability distribution functions would still be of different forms; in other words, the discrepancy is not due to the geometrical conditions imposed by the point-injection, radial diffusion arrangenment used in the present work.

Appendix D. Calibration of Flow Manometer and Colorimetric Standards.

Flow Manometer. Water was run through the apparatus and through the manometer until it was certain that no air was left in the tubes. The constant-head tank was allowed to fill until overflow was constant; the flow-rate was then adjusted to a chosen value by the control-valve. A reading of the pressure-drop across the orifice was then taken, and the flow-rate obtained by timing the collection of a measured volume (about 10,000 cc) of water as it over flowed from the tower. A second reading of the manometer was then taken; if it differed from the first, the two were averaged. When the volumetric rate of flow was plotted against the manometer head on log-log paper, the slope was exactly 1/2. The plot was is shown in Graph 22.

Colorimetric Standards. A portion of the original indicator solution made up (24.80 gms/litre) was diluted 1,000 times, and used as a reference standard (referred to in the lab. notes as $^{s}/1000$). Methylene blue solutions of various other known concentrations were then compared.with the $^{s}/1000$ in the Duboscq colorimeter. The color-densities of the various solutions were plotted against their concentrations, thus giving a curve from which the concentration of any solution of known color-density could be determined. It was noticed that it was advisable to compare solutions which were as close as possible in concentration, as the color appeared to vary with the concentration. Tap-water was used as the diluent throughout. The color-density/concentration curves are given in graphs 20 and 21.

Appendix E Addittional Notes on Experimental Technique.

Packings. Most of the work was done with 4-mm. and 6-mm. glass beads. These were cleaned with chromic acid before starting the experiments to discourage air-bubbles from clinging to them.

It is necessary to treat the packing in a standardised way in order to keep the free volume constant. In this work, the glass beads were stirred under water with a thin rod after every run, and measurements of free volume and pressure-gradient were made with the packing in a similar state of consolidation.

The 3/8-in. (nOminal diameter) spheres and ¼-in. Berl saddles could not be stirred in this way. They were dumped into the tower when it was full of water and lightly tamped down.

The alumina spheres as received were not uniform in shape or size., so they were sorted by hand to remove the more obviously deformed or outsize ones. The volume of a known number of the sorted spheres was then measured by displacement, and the mean diameter calculated. It was found to be 11 mm. (0.43 in) which is not very close to the nominal diameter of 3/8 (0.375) inch.

Methylene Blue Solutions. It was found advisable to warm the solution when making it up, to make certain that all the dye dissolved. It dissolves slowly, and it is difficult to see whether any solid is left. The exact concentration of the indicator solution need not be known, since it does not enter into the calculation. A solution containing 25 - 30 gms/litre was used; this approaching saturation, and as will be explained later, the highest practicable concentration was required.

A concentration of about 1/1000 of this was found to be the easiest to estimate accurately and quickly. When less than $\frac{1}{4}$ of this concentration, the accuracy of the determination is poor, and when more than 2 or 3 times this, the sample must be diuted to give accurate results.

For a superficial flow velocity $v = 1$ cm/sec, using a packing of spheres, the true mean Linear velocity through the packing is about 3 cm/sec., & the the volumetric rate of flow 70 cc/sec. Since the diameter of the injector tube was approximately .08 cm., injection of indicator solution at the true linear flow, velocity (as was originally considered desirable) would give an effluent solution with a concentration equal to

$$\frac{3 \times \pi \times (\cdot 04)^2}{70}\text{; or about } \cdot 002 \text{ of the}$$

indicator solution. Since the concentration of the effluent gives an idea of the range of concentration of the samples which will have to be analyzed, and since the concentration of the methylene blue solution can not be greatly increased, it is necessary to inject the indicator solution at a higher velocity (say 2-5 times as fast) than that of the main stream if samples of a suitable range of concentration are to be obtained. No significant variation in the observable results was noticed when the injection rate was varied, so that the procedure was probably permissible.

Apparatus. The nanometers used to meter the flow are shown in Fig. VII. There was a tendency for air to collect in the tubes leading from the metering orifice to the namometer, so the latter were so constructed ~~as~~ to allow water to be run through them and flush out air before readings were taken. In the case of the water manometer, used for flow rates up to about 150 cc/se., water was allowed to run out of the right-hand limb, the lower stop-cock was then closed, and the puffer used to force the water levels over into the two left-hand limbs (slugs of air which formed in the left-hand limbs in the process could be removed by raising and lowering the levels by alternately puffing in air and allowing it to escape). The upper stop-cock was then closed, to prevent air being sucked in through the puffer by the negative pressure developed beneath the metering orifice when the water valve was wide open. The mercury manometer, used only at the highest attainable flow-rates, was so designed that when the mercury reservoir was lowered, water could be run through both limbs and out of the lower stop-cock.

The steel capillary tubes used to collect the samples were very flexible, and in order to fix the position of their open ends with certainty it was necessary to attach them to a rigid frame, which is shown in Fig. IX. It was considered desirable for the liquid leaving the top of the tower to overflow symmetrically, at any rate at low flow-velocities, in order not to distort the flow in the tower. To ensure this, a 6-inch high metal extension was attached to the top of the glass tower, and 6 equally-spaced 1/10 inch holes were drilled in it, in a horizontal plane some 4 inches from the top. While at higher rates of flow the water overflowed at the top of this extension-piece, at lower rates it flowed exclusively through the six holes; the flow through each was equal and practically unaffected by any small tilt of the tower, because this would produce only a small fractional change in the head above any one hole.

The connections between the metal container at the bottom of the tower, the tower itself, and the extension piece at the top were secured and made leak-proof by the use of rim cement, and by a plastic waterproofing material which was forced tightly into the joint after assembly.

APPENDIX F. SAMPLE CALCULATION

Analysis of Runs 76 - 83 inclusive. All concentrations of the samples are expressed, as was the general case throughout, with reference to a standard solution of $\frac{1}{1000}$ the concentration of the original indicator solution injected. The concentrations of the effluent and the samples were found as outlined in Section III (Procedure).

The general statement of the probability expression used throughout these analyses is of the form

$$\frac{Cx}{C^*} = \frac{V}{4\pi FD} e^{-r^2 v/4FDx}$$

where

C Concentration of sample collected

C* Concentration of effluent liquid

x Axial distance between the injector capillary and sampling plane

V Volume of water flow c.c./sec.

F Free volume of the packing

D Diffusion coefficient Cms ^{2}/sec.

r Radial sampling distance from axis of the tower

v Superficial flow velocity of water. Cms/sec.

The general approach is to plot $\text{Log}_{10} \frac{Cx}{C^*}$ vs. r^2/x when the slope of the line is given by $-\frac{v}{9.2 FD}$ and the intercept by $\text{Log}_{10} \frac{V}{4\pi FD}$. The best line is drawn through the points obtained, consistent with satisfying the equation above. Upon averaging the individual results of Runs 76-83, Table A is obtained. Table B shows the results for the individual runs.

TABLE A

Run Numbers	Tube Number	$\frac{Cx}{C^*}$ (average)	Log $\frac{Cx}{C^*}$ (average)	$\frac{r^2}{x}$	Volumetric rate of flow (c.c./sec)	Superficial flow velocity CMS/sec
	1	288	2.460	0		
76-83	2	224	2.350	0.0113	21.7	0.0314
	3	143	2.156	0.064		
	4	60.2	1.780	0.129		
	5	10.8	1.034	0.225		

A plot is made of Log $\frac{C_x}{C^*}$ (average) vs. $\frac{r^2}{x}$

It is seen that the slope of the line on the plot $= -\frac{v}{9.2\ FD} = -5.75$

or substituting $FD = +\frac{0.314}{9.2 \times 5.75} = 0.00594$

But the intercept on the graph at $\frac{r^2}{x} = 0$ is given by $\log_{10} \frac{V}{4\pi FD}$

Intercept $= 2.464 = \log_{10} \frac{V}{4\pi\ FD}$ where V is the volume of flow /sec.cc.

Then $291 = \frac{21.7}{4\pi\ FD}$

$\therefore FD = 0.00594$

i.e. the values of F D as calculated from the slope and the intercept on the plot, give the same value for F D .

$$\text{Hence } D = \frac{.00594}{F} = \frac{.00594}{0.36} = 0.0165 \text{ cm}^2/\text{sec}$$

Centre-Line Sampling Method. In some cases samples were taken on the centre-line of the tower only. Then: $\frac{C_x}{C^*} = \frac{V}{4\pi FD}$ thus, for a given value of V, it is only necessary to average the values of $\frac{C_x}{C^*}$ for all the samples to find the best value of FD.

Table B.

Run No:	Tube No:	C	x	C^*	$\frac{Cx}{C^*}$	r	$\frac{r^2}{x}$
76	1	0.795	23.5	0.051	364	0	0
	2	0.860			394	0.266	0.0113
	3	0.230			105	1.51	0.064
	4	0.110			50.5	3.05	0.129
	5	0.020			9.17	5.30	0.225
77	1	4.38	23.5	0.245	418	0	0
	2	5.15			492	0.266	0.0113
	3	3.70			354	1.51	0.064
	4	1.45			138	3.05	0.129
	5	0.44			42	5.30	0.225
78	1	7.90	23.5	0.675	275	0	0
	2	5.10			178	0.266	0.0113
	3	3.20			112	1.51	0.064
	4	2.63			91.6	3.05	0.129
	5	0.187			6.52	5.30	0.225
79	1	2.15	23.5	0.46	110	0	0
	2	1.93			98	0.266	0.0113
	3	1.68			86	1.51	0.064
	4	0.63			32.2	3.05	0.124
	5	0.105			5.37	5.30	0.225
80	1	5.30	23.5	0.368	340	0	0
	2	2.40			154	0.266	0.0113
	3	1.15			74	1.51	0.064
	4	0.51			32.7	3.05	0.129
	5	0.035			2.24	5.30	0.225
81	1	5.07	23.5	0.255	470	0	0
	2	2.50			231	0.266	0.0113
	3	1.62			150	1.51	0.064
	4	0.24			22.2	3.05	0.129
	5	0.015			1.39	5.30	0.225
82	1	1.072	23.5	0.205	124	0	0
	2	0.63			73	0.266	0.0113
	3	0.060			6.95	1.51	0.064
	4	0.020			2.34	3.05	0.129
	5	-----			-----	-----	-----
83	1	1.17	23.5	0.135	216	0	0
	2	0.995			176	0.266	0.0113
	3	1.07			189	1.51	0.064
	4	0.545			96.3	3.05	0.129
	5	0.09			15.9	5.30	0.225

Appendix H. Nomenclature, definitions and symbols.

A	cross-sectional area of tower - sq. cms.
A_f	factor to be used in fanning equation to correct for variations in ratio of particle diameter to tower diameter.
$\bar{C}$	root mean square velocity of particle in random motion (in 3 dimensions).
C	concentration of sample taken at point (x , r) ---1000 x(cc. of injected indicator solution per cc. of sample). (i.e relative to 5/1000 standard)
C^*	mean concentration of total stream passing through tower-same units as C.
d	particle diameter ---cms.
dp	nominal particle diameter (cms) for irregular shapes --eg. Berl saddles.
D	apparent diffusion co-efficient (sq. cms per sec.) as defined in Appendix C.
D'	co-efficient of molecular diffusion
E	eddy diffusion coefficient.
f	Fanning friction factor, as defined in Appendix B.
F	fraction of overall volume of packing which is not occupied by the substance of the packing particles.
L	depth (cms) (measured parallel to axis of tower) of packing through which pressure drop is ΔP, dynes per sq.cm., when superficial velocity is v cms. per sec.
n	number of molecules (defined by context.)
N	number of spheres in unit overall volume of packing.
ΔP	pressure-drop (dynes per sq.cm., unless otherwise stated) through depth L cms. of packing at superficial velocity v cms. per sec.
r	distance of sampling-points from axis of tower (cms).
t	time (secs.)
u	mean linear velocity (in direction parallel to tower axis) of fluid through interstices of packing - cms per sec.

v	$= {}^{V}\!/_{A} =$ volumetric rate of flow divided by cross-sectional area of tower, or superficial velocity of flow (cms. per sec.)
V	volumetric rate of flow, or volume of fluid flowing through tower in unit time --cc. per sec.
x	distance of cross-sectional plane in which sample is taken from that of injection-point-cms. (See Appendix C).
Z	collision frequency, or frequency with which particle in random motion changes direction.
ρ	density of fluid — gms/cc.
μ	viscosity of fluid — gms/sec.-cm.

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TABULATION OF RESULTS.

GRAPHS.

FIGURES.

RUN NUMBERS INCLUSIVE	PACKING TYPE AND DIAMETER	DIFFUSION COEFFICIENT D CMS ^{2}/sec	FREE VOLUME F.	FLOW VELOCITY CMS/SEC	AXIAL DISTANCE FROM INJECTOR TO SAMPLING PLANE CMS	$\frac{Cx}{C^*}$					Average value of $\frac{Cx}{C^*}$					Radial Sampling distance CMS				
						TUBE I central	TUBE II	TUBE III	TUBE IV	TUBE V	TUBE I	TUBE II	TUBE III	TUBE IV	TUBE V	TUBE I	TUBE II	TUBE III	TUBE IV	TUBE V
2 - 8	6 mm glass beads	0.082	0.36	1.07												0	1.03	2.06	3.14	4.3
2 (a)					10.25	160	107	10.9	---	---										
2 (b)					10.25	186	68	20.2	---	---										
3 (a)					10.25	130	49.5	6.3	---	---										
3 (b)					10.25	175	32.3	3.58	---	---										
4					10.25	128	188	81	49.9	---										
2 - 4											157	89	22.4	9.9	---					
5					20.5	156	131	47.5	4.87	----										
6					20.5	130	135	75.72	---	---										
7					20.5	188	166	37.4	12.1	----										
8					20.5	326	233	152.0	50.0	----										
2 - 8											200	158	78	16.6	---					
9 - 15	6 mm glass beads	0.196	0.36	1.66												0	1.03	2.06	3.14	4.3
9					10.6	102	62.5	15.9	0.63											
10					10.6	122	66.0	19.6	0.44	---										
11					10.6	164	55.2	6.15	---	---										
12					10.6	98	43.7	10.1	---	----										
9 - 12											121	55.6	12.9	0.53	---					
13					15.15	162	113	30.4	0.57	----										
14					15.15	129	140	34.2	0.95	----										
15					15.15	131	90.5	13.1	0.79	----										
13 - 15											141	81.1	25.9	0.77	---					

RUN NUMBERS INCLUSIVE	PACKING TYPE AND DIAMETER	DIFFUSION COEFFICIENT -D CMS^2/sec	FREE VOLUME F.	FLOW VELOCITY CMS/SEC	AXIAL DISTANCE FROM INJECTOR TO SAMPLING PLANE CMS	$\frac{Cx}{C^*}$					Average Value of $\frac{Cx}{C^*}$					Radial Sampling distance CMS				
						TUBE I central	TUBE II	TUBE III	TUBE IV	TUBE V	TUBE I	TUBE II	TUBE III	TUBE IV	TUBE V	TUBE I	TUBE II	TUBE III	TUBE IV	TUBE V
16 - 23	6 mm glass beads	0.225	0.36	1.85												0	1.03	2.06	3.14	4.3
16					12.9	76.6	62.5	15.9	3.64	1.01										
17						57.0	12.1	3.39	0.645	---										
18						99.5	59.0	17.3	0.555	---										
19						167	152	19.8	1.27	---										
16 - 19											100	71.4	14.1	1.55	---					
20					9.75	163	71.9	10.0	1.03	---										
21						103	42.6	16.6	0.545	---										
22						96.9	120	5.2	0.73	---										
23						131	70.5	6.8	2.1	---										
20 - 23											126	76	9.6	1.11	---					
24 - 31	6 mm glass beads	0.0103	0.36	0.25												0	0.535	1.08	1.66	2.26
24					12.3	352	244	46.2	0.79	---										
25						216	72.2	22.9	10.6	---										
26						297.5	124	38.9	1.4	---										
27						141.5	124	79.0	2.66	---										
24 - 27											249	200	40.1	3.92	---					
28					17.15	411	291	55.7	4.3	---										
29						377	230	129	---	---										
30						393	215	40.0	9.3	---										
31						361	244	222	21.0	---										
24 - 31											387	246	112	8.75						
32 - 40	6 mm glass beads	0.020	0.36	0.32																
32					17.2	318	160	25.9	---	---										
33						332	151	100.0	---	----										
34						254	258	95	38.7	---										
35						241	183	99.0	49.5	---										
36						172	485	106	35	---										

RUN NUMBERS INCLUSIVE	PACKING TYPE AND DIAMETER	DIFFUSION COEFFICIENT D CMS ^{2}/sec	FREE VOLUME F	FLOW VELOCITY CMS/SEC	AXIAL DISTANCES FROM INJECTOR TO SAMPLING PLANE CMS	$\frac{Cx}{C^*}$					Average value of $\frac{Cx}{C^*}$					Radial Sampling distance CMS				
						TUBE I central	TUBE II	TUBE III	TUBE IV	TUBE V	TUBE I	TUBE II	TUBE III	TUBE IV	TUBE V	TUBE I	TUBE II	TUBE III	TUBE IV	TUBE V
32 - 36											266	158	85	41.5						
37					13.65	404	268	67.3	---	----										
38					13.65	240	112	67.7	----	----										
39					13.65	87	112	116	37.4	----										
40					13.65	93	156	105	2.0	16.8										
38 - 40											206	162	79	19.7	4.2					
41 - 49	4 mm glass beads	0.0105	0.36	0.28												0	0.535	1.08	1.66	2.26
41					17.35	225	238	48.0	5.8	---										
42						360	218	105.4	6.3	---										
43						338	510	300	39.9	---										
44						423	260	130	3.25	---										
45						424	384	72	45	---										
41 - 45											352	314	131	20.0	--					
46					21.9	221	294	199	33.2	---										
47					21.9	405	306	79.5	10.3	---										
48					21.9	242	127	63.5	14.9											
49					21.9	536	333	120	68.0											
46 - 49											376	265	113	31.6						

RUN NUMBERS INCLUSIVE	PACKING TYPE AND DIAMETER	DIFFUSION COEFFICIENT D CMS ^{2}/sec	FREE VOLUME F	FLOW VELOCITY CMS/SEC	AXIAL DISTANCE FROM INJECTOR TO SAMPLING PLANE CMS	$\frac{C\chi}{C^*}$					Average value of $\frac{C\chi}{C^*}$					Radial Sampling distance CMS				
						TUBE I central	TUBE II	TUBE III	TUBE IV	TUBE V	TUBE I	TUBE II	TUBE III	TUBE IV	TUBE V	TUBE I	TUBE II	TUBE III	TUBE IV	TUBE V
50 - 57	4 mm glass beads	0.033	0.36	0.69							375	282	153	37.5	13.4	0	0.535	1.08	1.66	2.26
50					18.2	383	450	220	43.3	---										
51					18.2	262	116	13.5	---	---										
52					18.2	330	234	214	66	11.6										
53					18.2	364	405	290	35.2	9.26										
54					18.2	277	196	60.0	29	5.5										
55					18.2	528	107	64.5	21.2	1.9										
56					18.2	396	211	120	35.3	6.47										
57					18.2	480	521	217.2	71.0	73.0										
59 - 66	4 mm glass beads	0.133	0.36	1.75												0	1.11	2.14	3.34	4.27
59					16.05	133	152	35	3.52	---										
60					16.05	302	123	8.025	---	---										
61					16.05	201	66.4	4.69	---	---										
62					16.05	212	51.2	44.1	---	---										
59 - 62											212	98	15.0	---	---					
63					22.35	202	89	24.4	1.69	---										
64					22.35	214	140	30.1	6.65	---										
65					22.35	176	104	30.5	---	---										
66					22.35	160	45.7	42.1	4.1	---										
63 - 66											188	94.5	31.8	3.16	----					
67																				
		Experimental	runs	For trial	purposes only.															
68																				
69 - 75	4 mm glass beads	0.0812	0.36	1.22				CENTER LINE SAMPLING										CENTER LINE SAMPLING		
69					4.4	139														
70					6.7	155														
71					8.95	188														
72					16.30	259														
73					18.90	248														
74					21.6	273														
75					18.1	210														

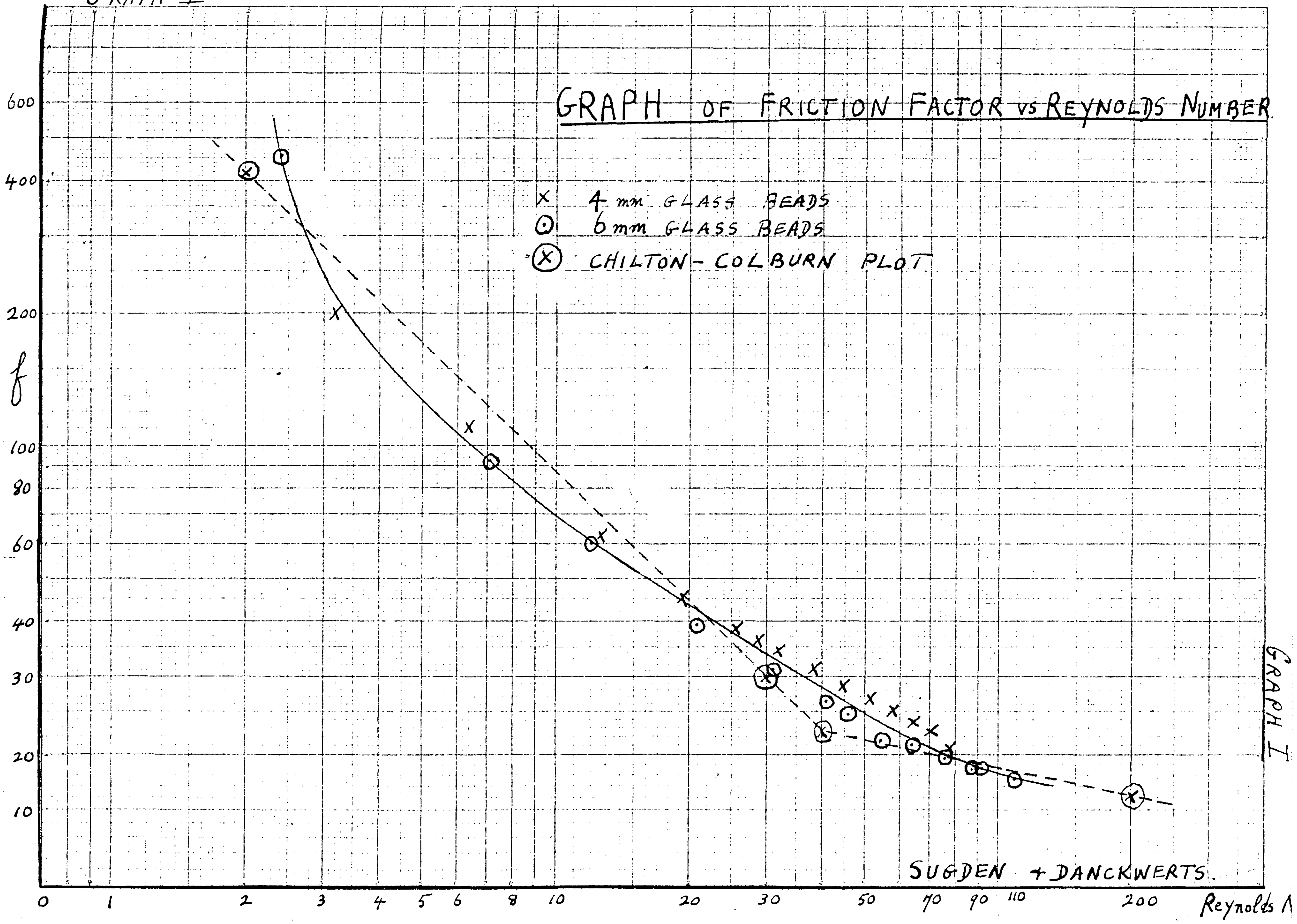
RUN NUMBERS INCLUSIVE	PACKING TYPE AND DIAMETER	DIFFUSION COEFFICIENT D CMS $^2/$sec	FREE VOLUME F	FLOW VELOCITY CMS/SEC	AXIAL DISTANCE FROM INJECTOR TO SAMPLING PLANE CMS	$\frac{Cx}{C^*}$					Average value of $\frac{Cx}{C^*}$					Radial Sampling Distance (cms.)
						TUBE I central	TUBE II	TUBE III	TUBE IV	TUBE V	TUBE I	TUBE II	TUBE III	TUBE IV	TUBE V	
92 - 103 (cont)																
101					22.60	72.5										
102					22.55	79.8										
103					22.40	101.5										
104 - 115	4 mm glass beads	0.224	0.36	2.36	----	CENTRE LINE SAMPLING					160					CENTER LINE SAMPLING
104					22.0	175										
105					22.1	183										
106					21.9	197										
107					22.1	179										
108					22.2	147										
109					22.1	167										
110					22.0	162										
111					22.0	130										
112					22.0	215										
113					21.9	111										
114					22.0	129										
115					22.2	123										CENTER LINE SAMPLING
116 - 121	1/4 inch Berl saddles	0.354	0.59	1.91	----	---	CENTRE LINE SAMPLING				50.2					
116					13.3	54.3										
117					13.5	34.0										
118					13.5	55.3										
119					13.2	71.2										
120					12.7	38.8										
121					13.1	47.8										

RUN NUMBERS INCLUSIVE	PACKING TYPE AND DIAMETER	DIFFUSION COEFFICIENT D CMS 2/sec	FREE VOLUME F	FLOW VELOCITY CMS/SEC	AXIAL DISTANCE FROM INJECTOR TO SAMPLING PLANE CMS	Cx/C*					Average value of Cx/C*					Radial Sampling Distance CMS				
						TUBE I central	TUBE II	TUBE III	TUBE IV	TUBE V	TUBE I	TUBE II	TUBE III	TUBE IV	TUBE V	TUBE I	TUBE II	TUBE III	TUBE IV	TUBE V
72 - 75											246									
76 - 83	6 mm glass beads	0.0165	0.36	0.314	23.5						288	224	143.0	60.2	10.8	0	0.515	1.22	1.74	2.29
76						364	394	105	50.5	9.17										
77						418	492	354	138	42										
78						275	178	178	92	6.5										
79						110	98	86	32	5.4										
80						340	154	74	33	2.24										
81						470	231	150	22.2	1.39										
82						124	73	7	2.34	---										
83						216	176	189	96.3	15.9										
84 - 91	4 mm glass beads	0.172	0.36	2.26	20.6						169	126	13.08	1.3	---	0	1.08	2.2	3.35	4.11
84						182	93	15.9	1.13	---										
85						130	77	7.22	---	---										
86						296	124	24.7	---	---										
87						138	148.6	4.16	---	---										
88						135	70.5	6.30	---	---										
89						114	116	5.40	---	---										
90						184	171	16.25	7.47	---										
91						173	208	25.0	---	---								CENTRE LINE SAMPLING		
92 - 103	6 mm glass beads	0.389	0.36	2.30	----		CENTRE LINE SAMPLING		90											
92					22.5	93.5														
93					22.5	76.5														
94					22.5	101														
95					22.7	87.5														
96					22.75	91.0														
97					22.75	93.0														
98					22.45	94.5														
99					22.50	145														
100					22.70	85														

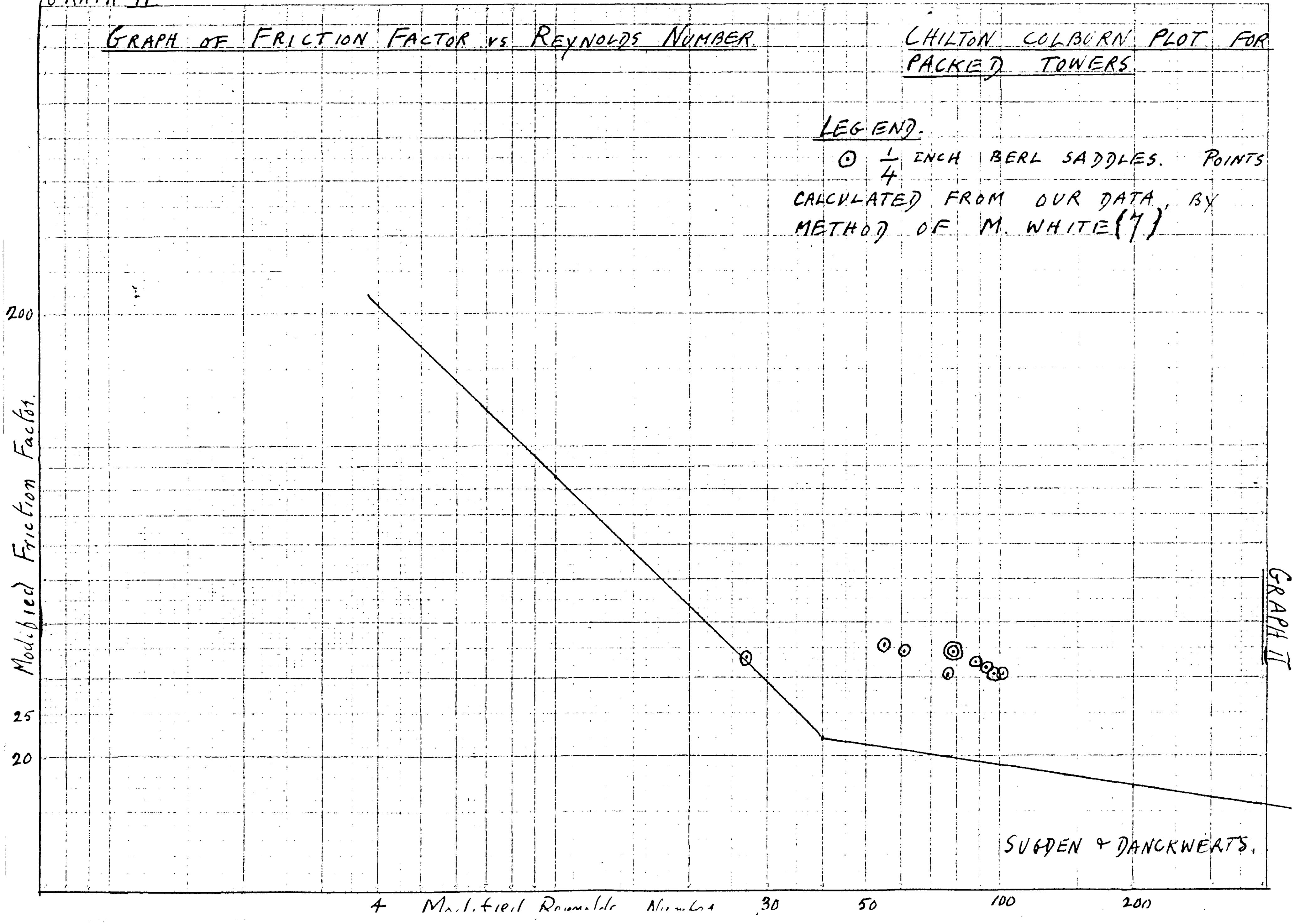
(continued)

RUN NUMBERS INCLUSIVE	PACKING TYPE AND DIAMETER	DIFFUSION COEFFICIENT D CMS ^{2}/sec	FREE VOLUME F.	FLOW VELOCITY CMS/SEC	AXIAL DISTANCE FROM INJECTOR TO SAMPLING PLANE CMS	$\frac{Cx}{C^*}$					Average value of $\frac{Cx}{C^*}$					Radial Sampling Distance (cms)					
						TUBE I	TUBE II	TUBE III	TUBE IV	TUBE V	TUBE I	TUBE II	TUBE III	TUBE IV	TUBE V						
122 - 130	$^3/_8$" Al_2O_3	0.200	0.40	1.057		CENTRE LINE SAMPLING					72.8						CENTRE LINE SAMPLING				
122	catalyst spheres				13.5	65.3															
123					13.8	64.8															
124					14.1	36.8															
125					13.5	73.5															
126					13.2	100															
127					14.0	43.4															
128					14.1	139															
129					14.0	58.0															
130					13.5	54.1															
131 - 142	$^3/_8$" Al_2O_3	0.227	0.40	1.057		CENTRE LINE SAMPLING					63.5						CENTRE LINE SAMPLING				
131	catalyst spheres				13.5	37.8															
132					14.2	57.8															
133					13.9	76.7															
134					13.9	62.2															
135					14.0	57.8															
136					14.2	50.2															
137					14.7	67.4															
138					13.75	94.5															
139					13.75	102															
140					13.50	55.4															
141					13.50	20.2															
142					13.50	50.0															

GRAPH I

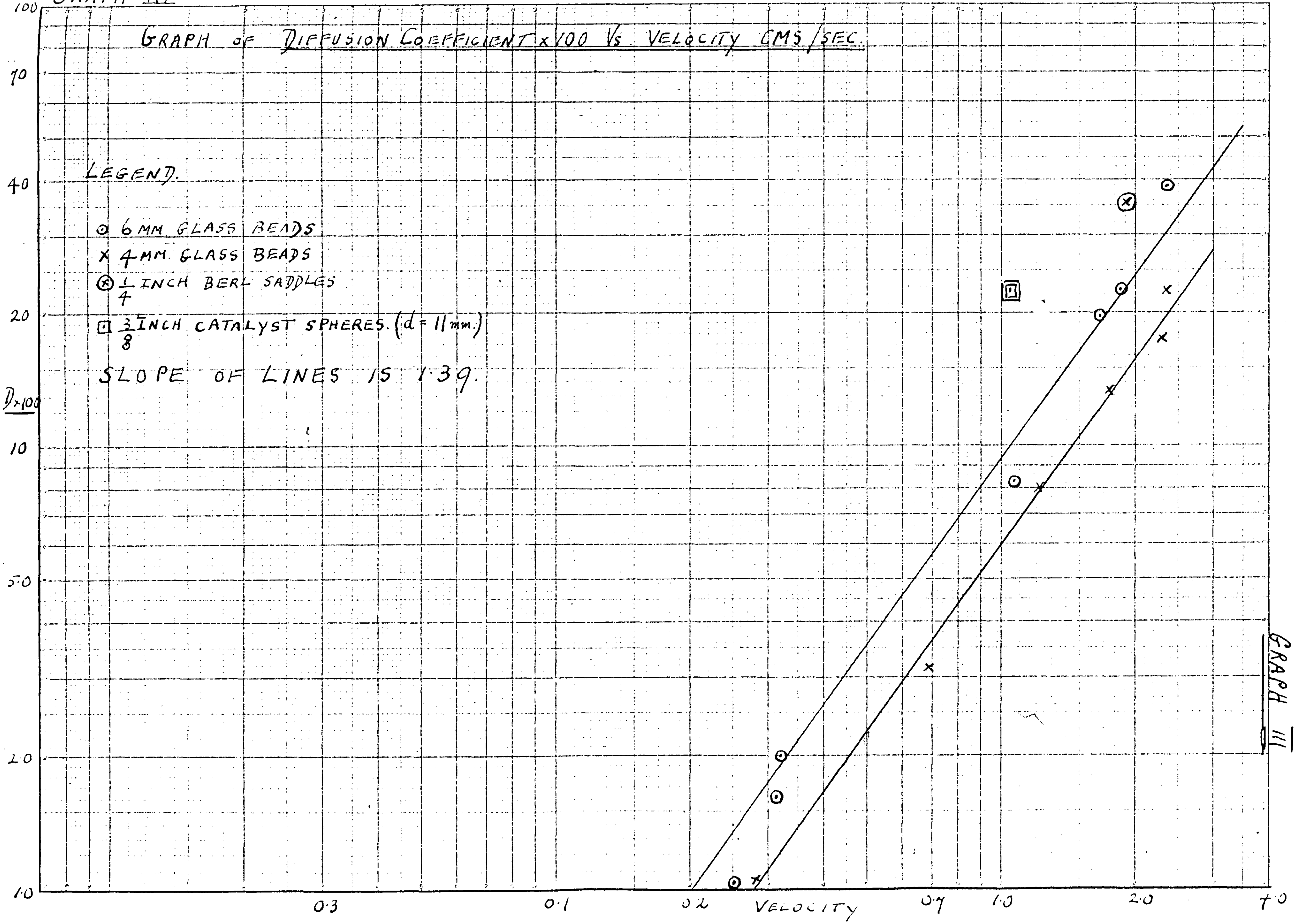

GRAPH OF FRICTION FACTOR vs REYNOLDS NUMBER
x 4 mm GLASS BEADS
⊙ 6 mm GLASS BEADS
⊗ CHILTON-COLBURN PLOT
f
600
400
200
100
80
60
40
30
20
10
0
1
2
3
4
5
6
8
10
20
30
50
70
90
110
200
Reynolds N
SUGDEN + DANCKWERTS.
GRAPH I

GRAPH II

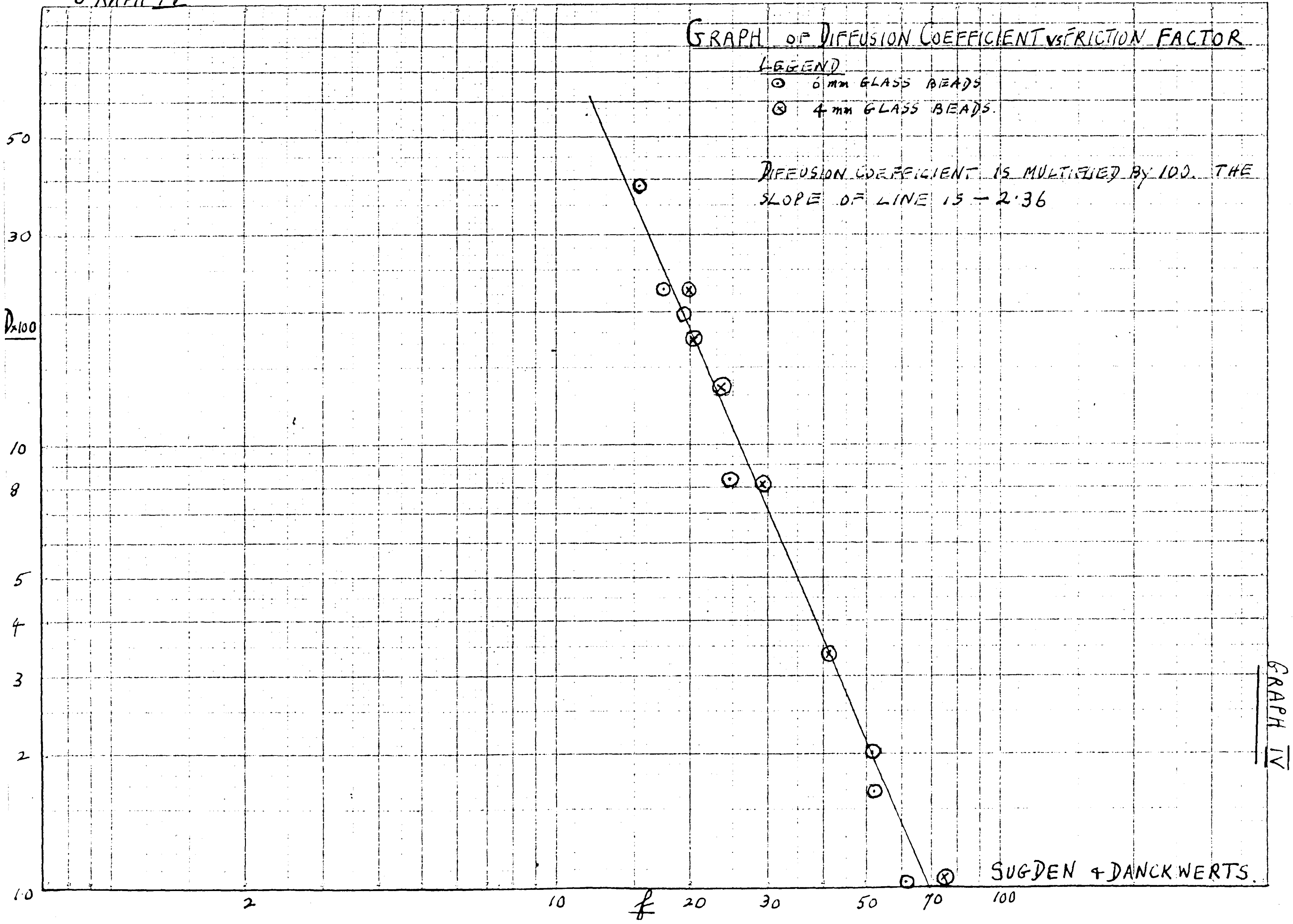


GRAPH II

GRAPH III


GRAPH OF DIFFUSION COEFFICIENT x 100 Vs VELOCITY CMS/SEC.
LEGEND.
⊙ 6 MM GLASS BEADS
x 4 MM. GLASS BEADS
⊗ 1/4 INCH BERL SADDLES
⊡ 3/8 INCH CATALYST SPHERES. (d = 11 mm.)
SLOPE OF LINES IS 1·39.
D x 100
100
70
40
20
10
5·0
2·0
1·0
0·3
0·1
0·2
VELOCITY
0·7
1·0
2·0
4·0
GRAPH III

GRAPH IV



GRAPH OF DIFFUSION COEFFICIENT vs FRICTION FACTOR

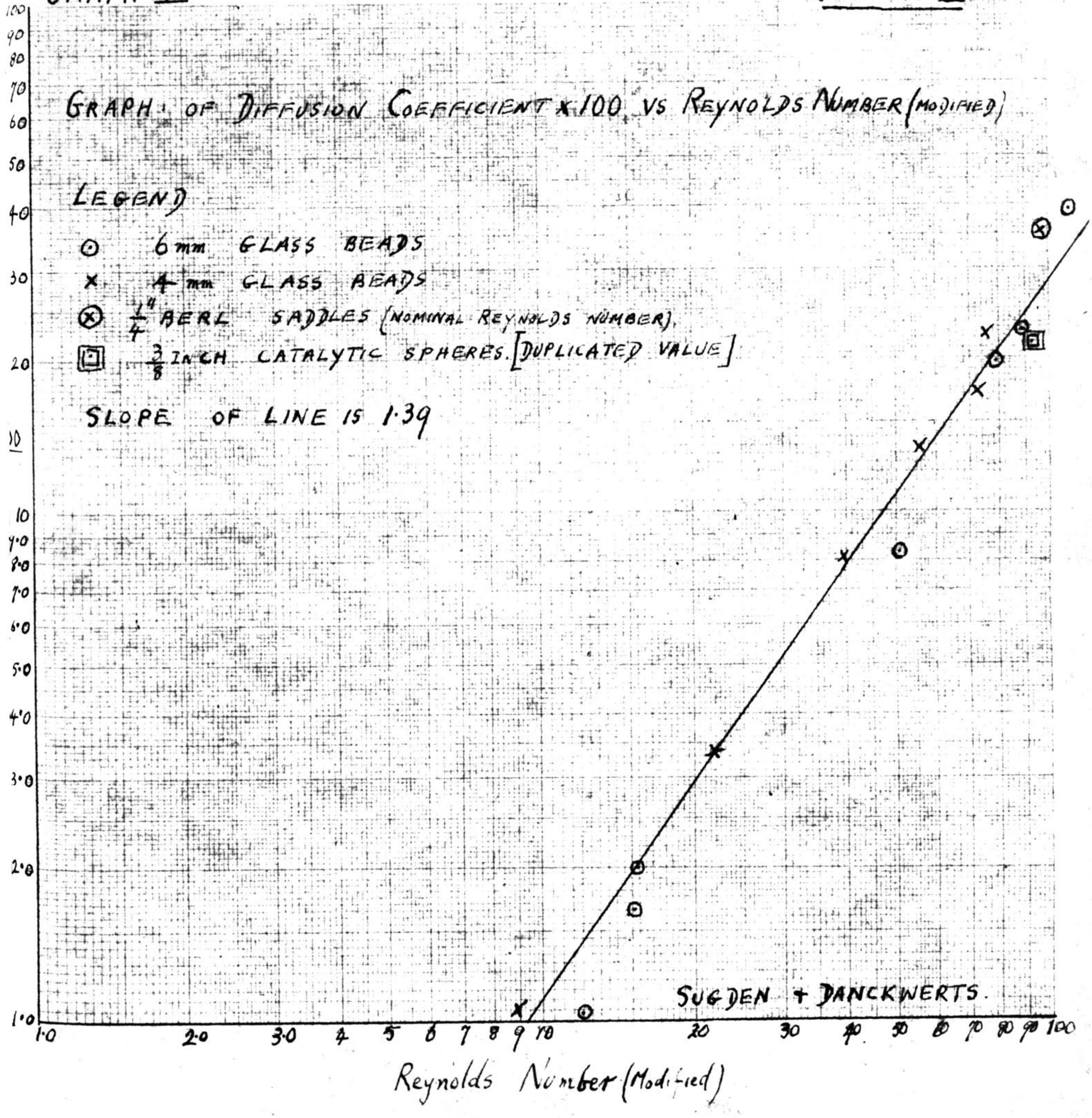
LEGEND
⊙ 6 mm GLASS BEADS
⊗ 4 mm GLASS BEADS.

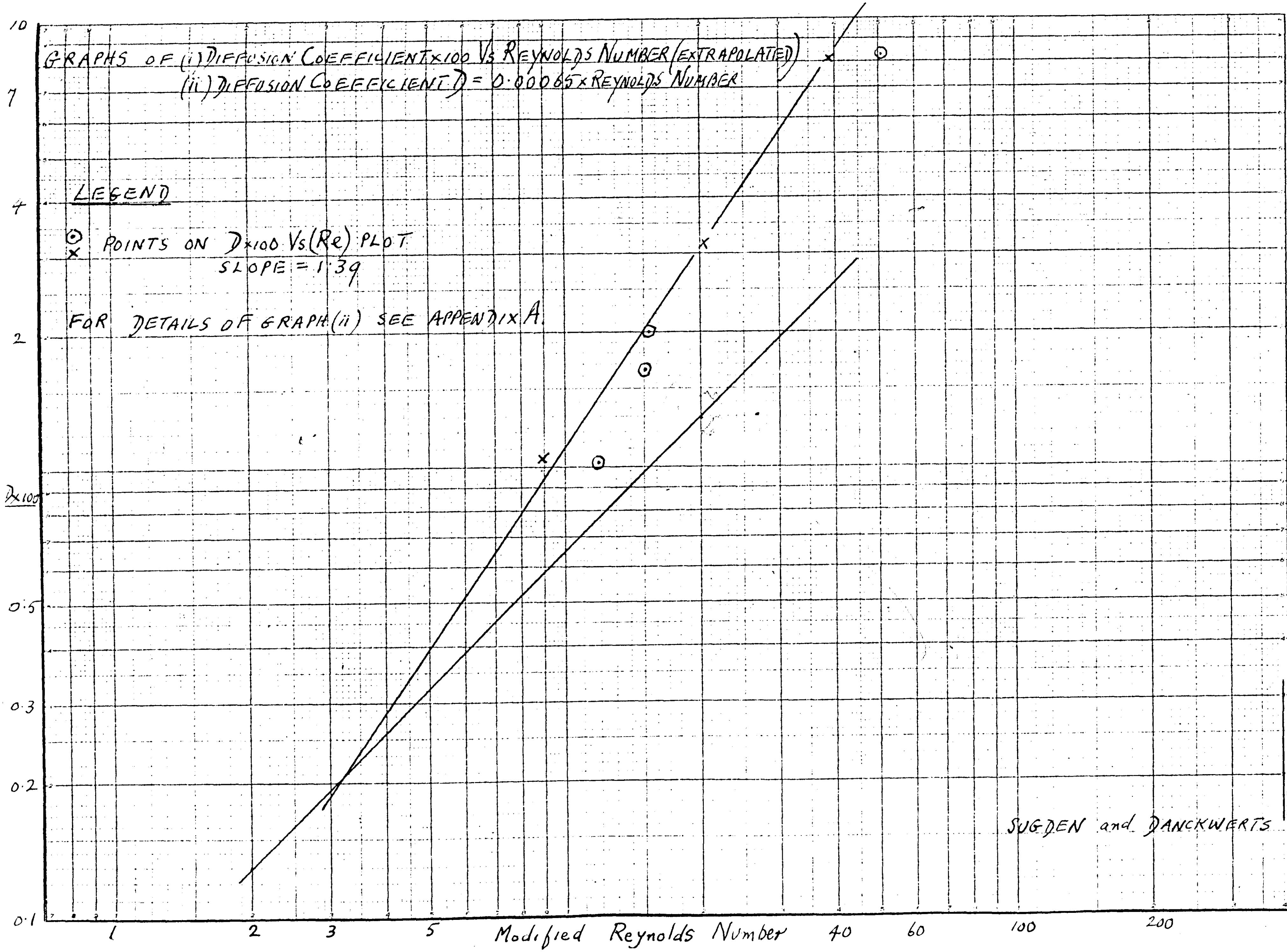
DIFFUSION COEFFICIENT IS MULTIPLIED BY 100. THE SLOPE OF LINE IS – 2·36

GRAPH IV

GRAPH V

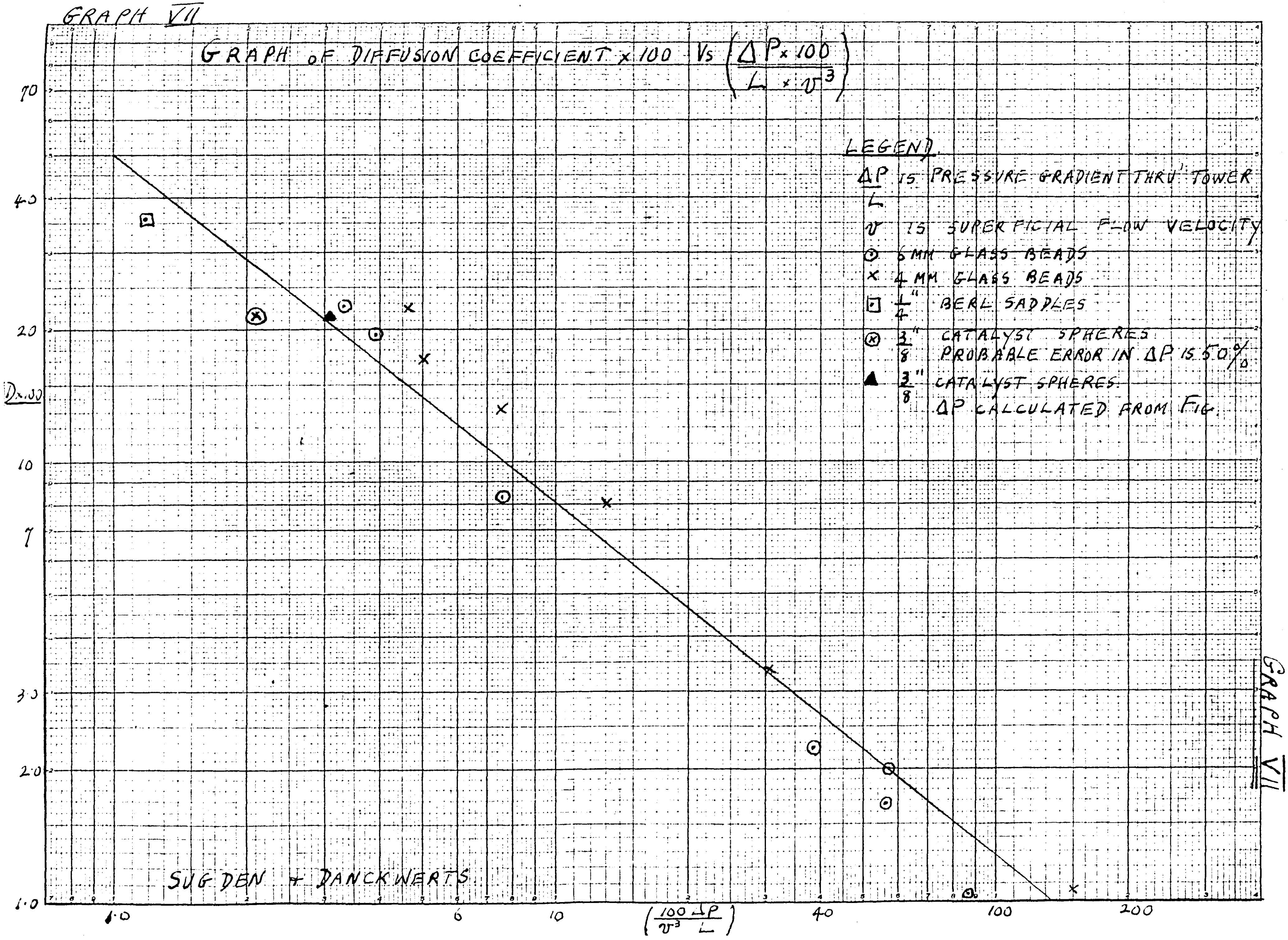
GRAPH V




GRAPHS OF (i) DIFFUSION COEFFICIENT x 100 Vs REYNOLDS NUMBER (EXTRAPOLATED)
(ii) DIFFUSION COEFFICIENT D = 0·00065 x REYNOLDS NUMBER
LEGEND
⊙ POINTS ON D x 100 Vs (Re) PLOT
x SLOPE = 1·39
FOR DETAILS OF GRAPH (ii) SEE APPENDIX A.
D x 100
10
7
4
2
0·5
0·3
0·2
0·1
1
2
3
5
40
60
100
200
Modified Reynolds Number
SUGDEN and DANCKWERTS

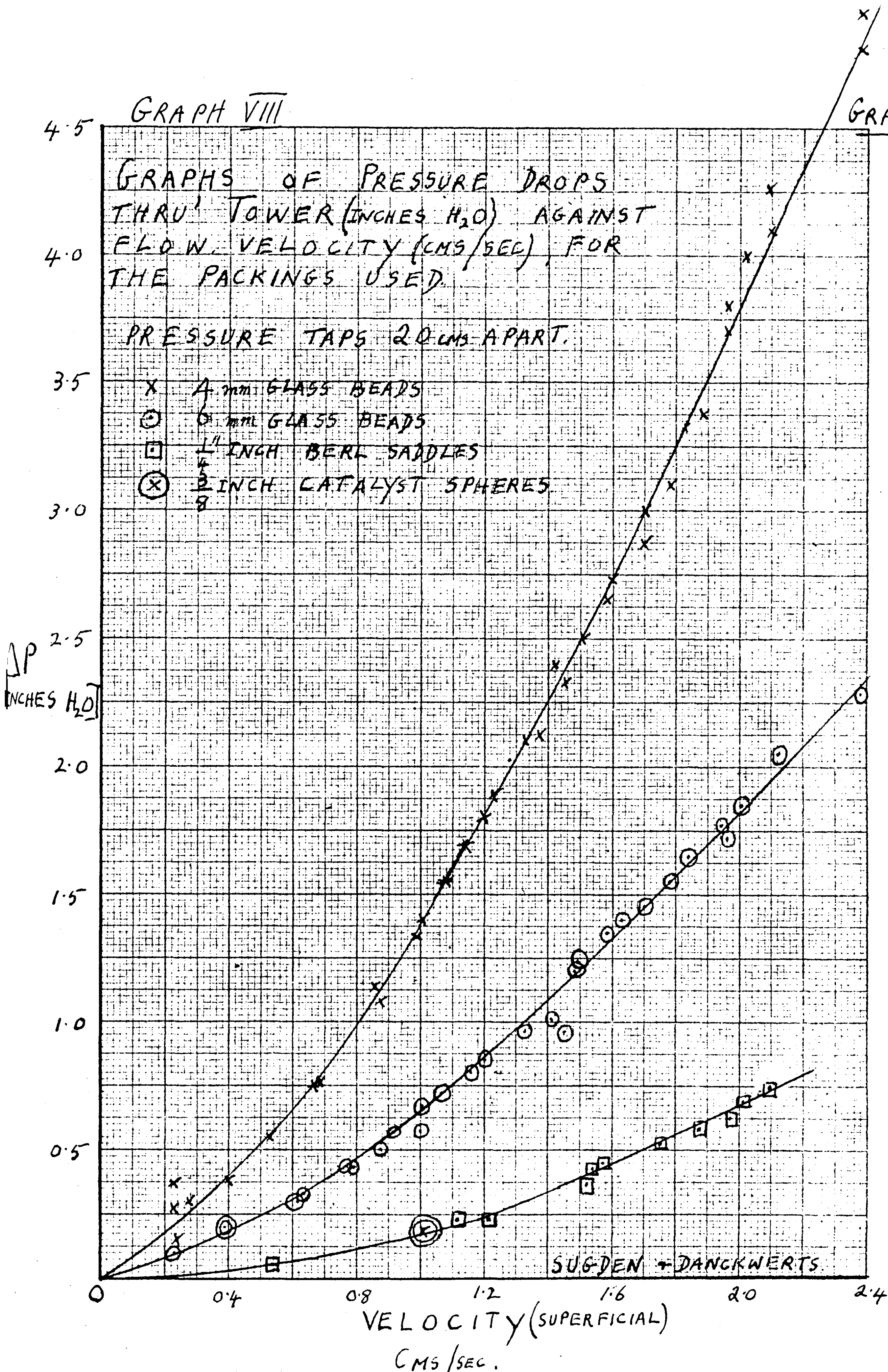
GRAPH VI

GRAPH VII

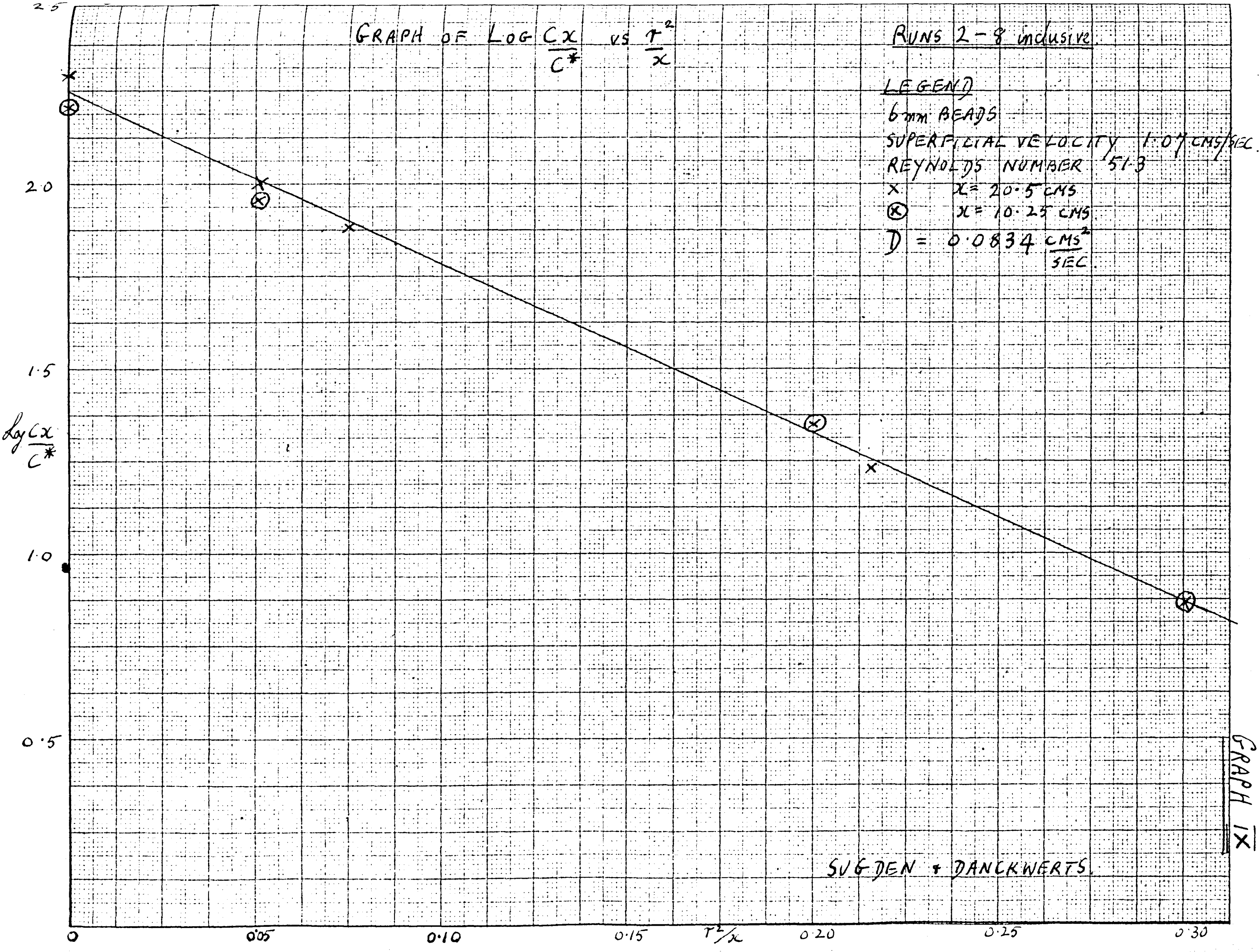


GRAPH VII

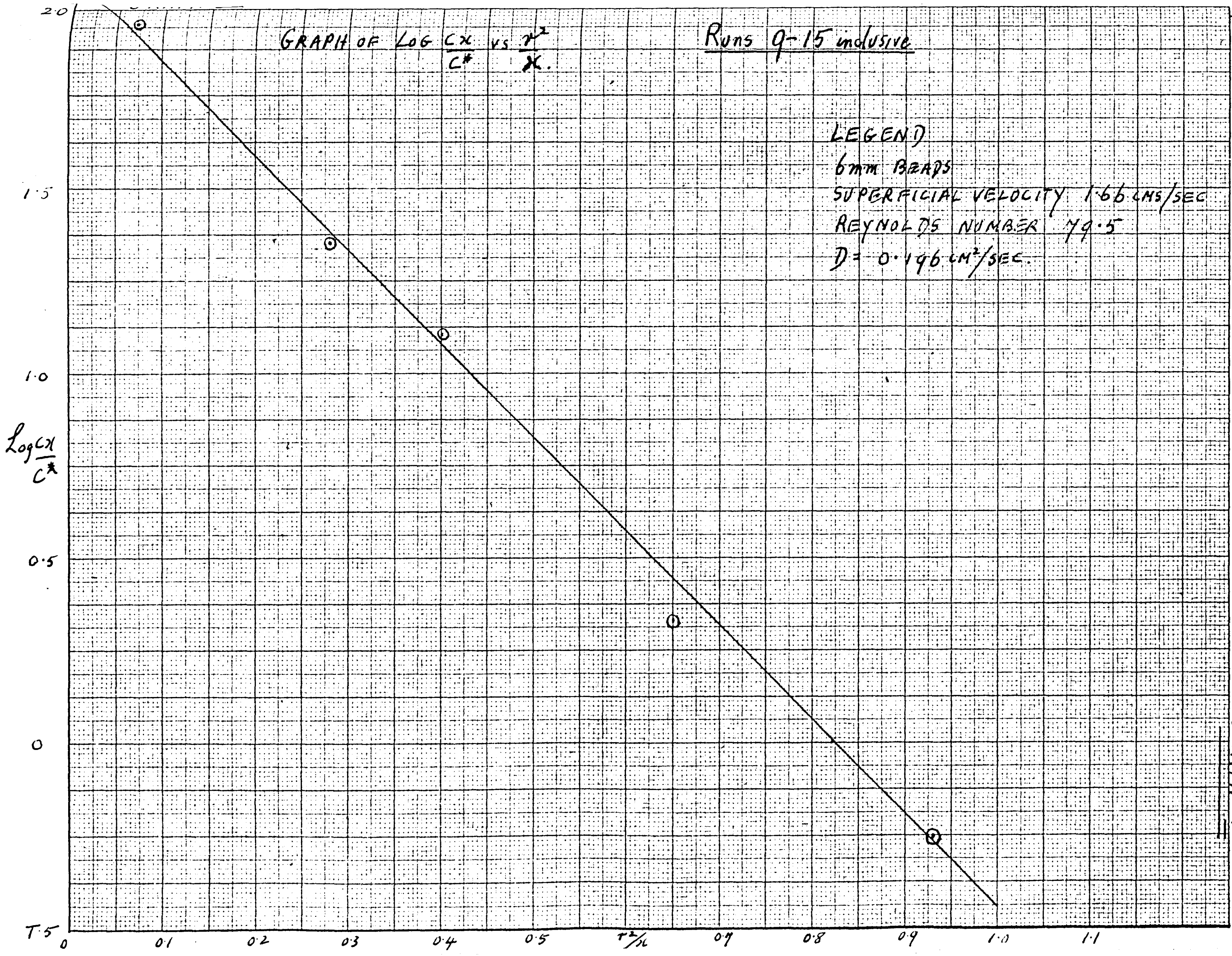
GRAPH VIII



GRAPH VIII

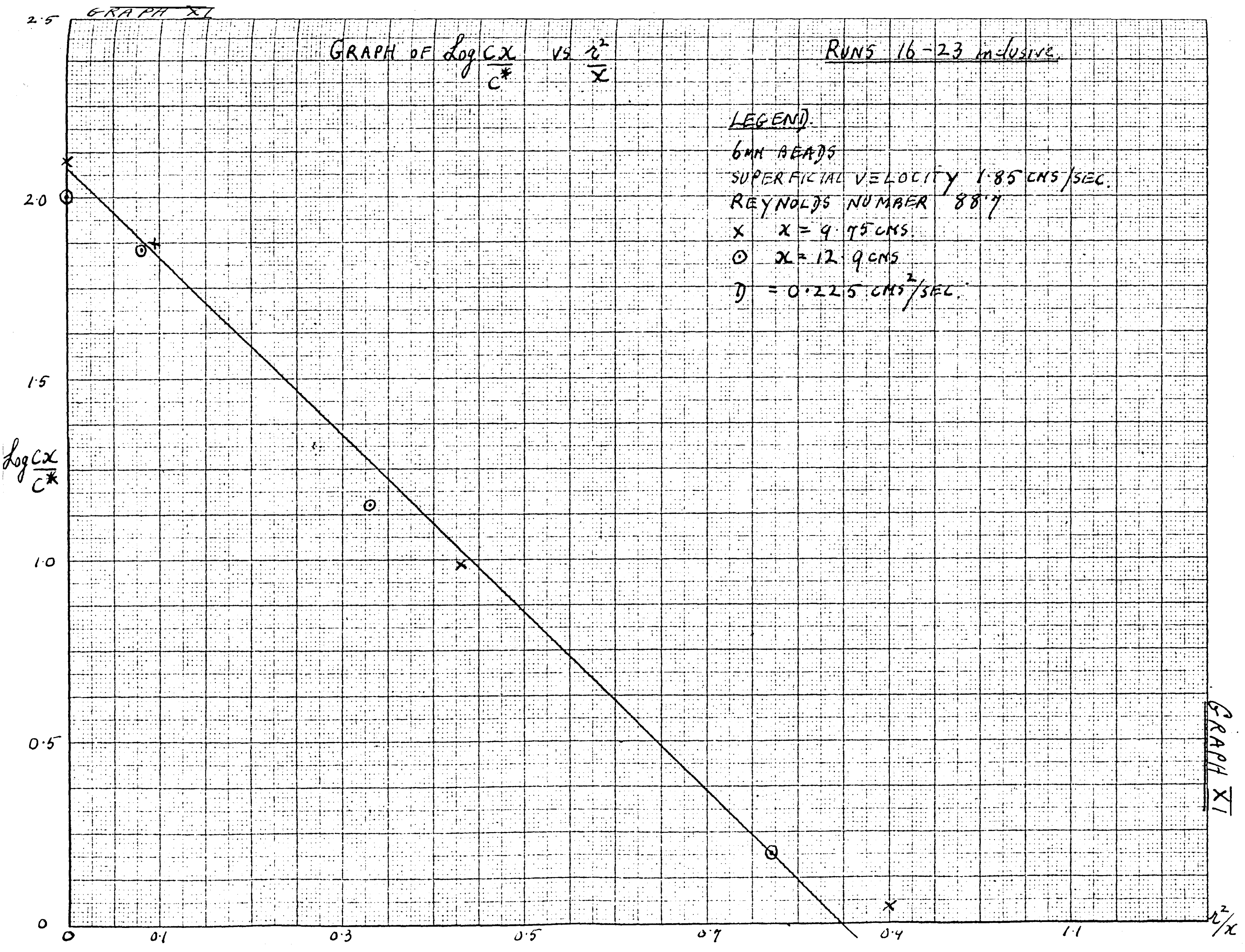
GRAPH OF LOG $\frac{Cx}{C^*}$ vs $\frac{r^2}{x}$
RUNS 2 – 8 inclusive
LEGEND
6 mm BEADS
SUPERFICIAL VELOCITY 1·07 CMS/SEC.
REYNOLDS NUMBER 51·3
× x = 20·5 CMS
⊗ x = 10·25 CMS
$D = 0·0834 \frac{CMS^2}{SEC}$
SUGDEN + DANCKWERTS.
2·5
2·0
1·5
1·0
0·5
$\log \frac{Cx}{C^*}$
0
0·05
0·10
0·15
r^2/x
0·20
0·25
0·30

GRAPH IX


GRAPH OF LOG $\frac{Cx}{C^*}$ vs $\frac{r^2}{x}$.
Runs 9-15 inclusive
LEGEND
6mm BEADS
SUPERFICIAL VELOCITY 1·66 CMS/SEC
REYNOLDS NUMBER 79·5
D = 0·196 CM²/SEC.
$\log \frac{Cx}{C^*}$
2·0
1·5
1·0
0·5
0
$\bar{1}$·5
0
0·1
0·2
0·3
0·4
0·5
r^2/x
0·7
0·8
0·9
1·0
1·1

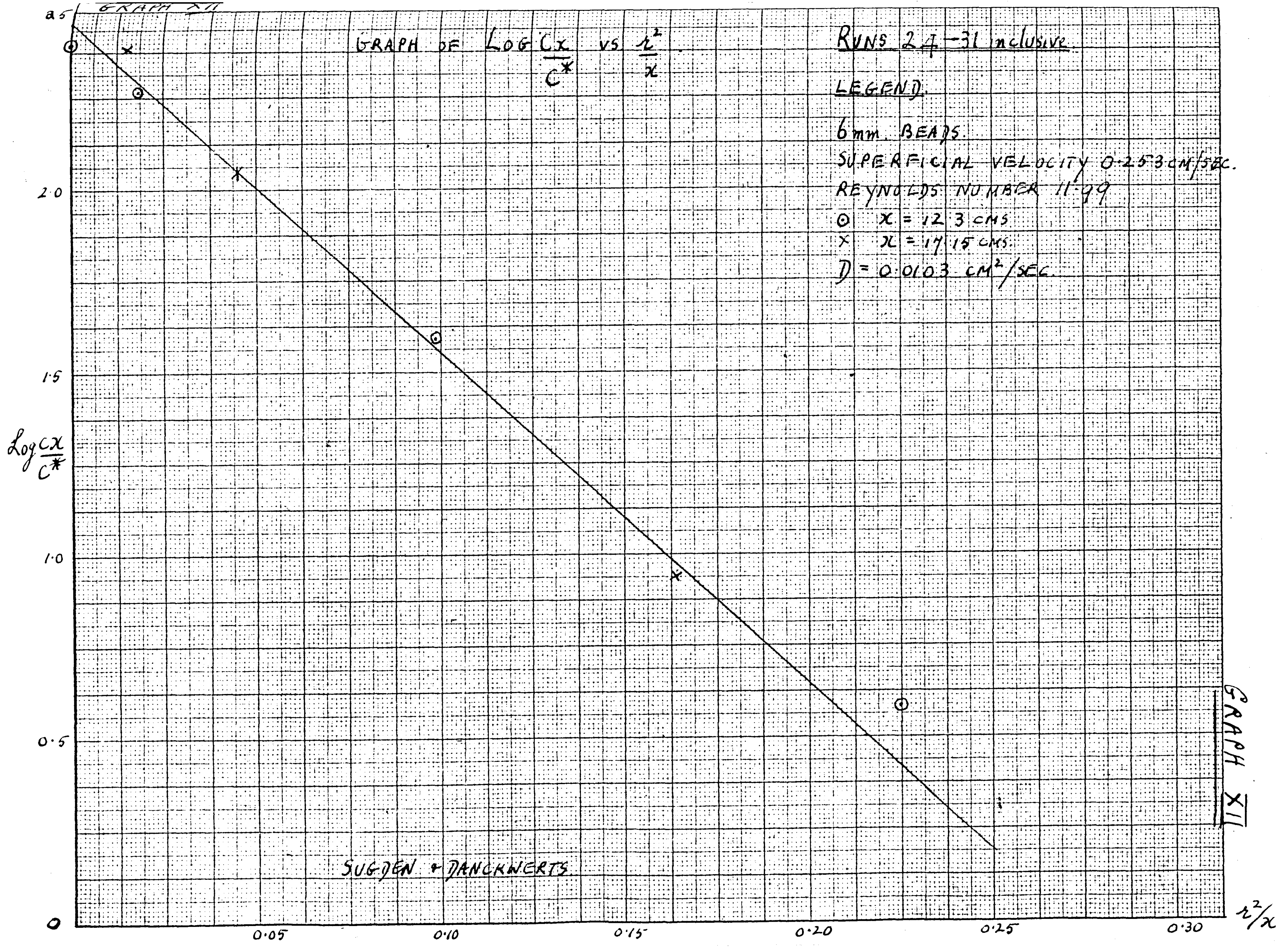
GRAPH XI

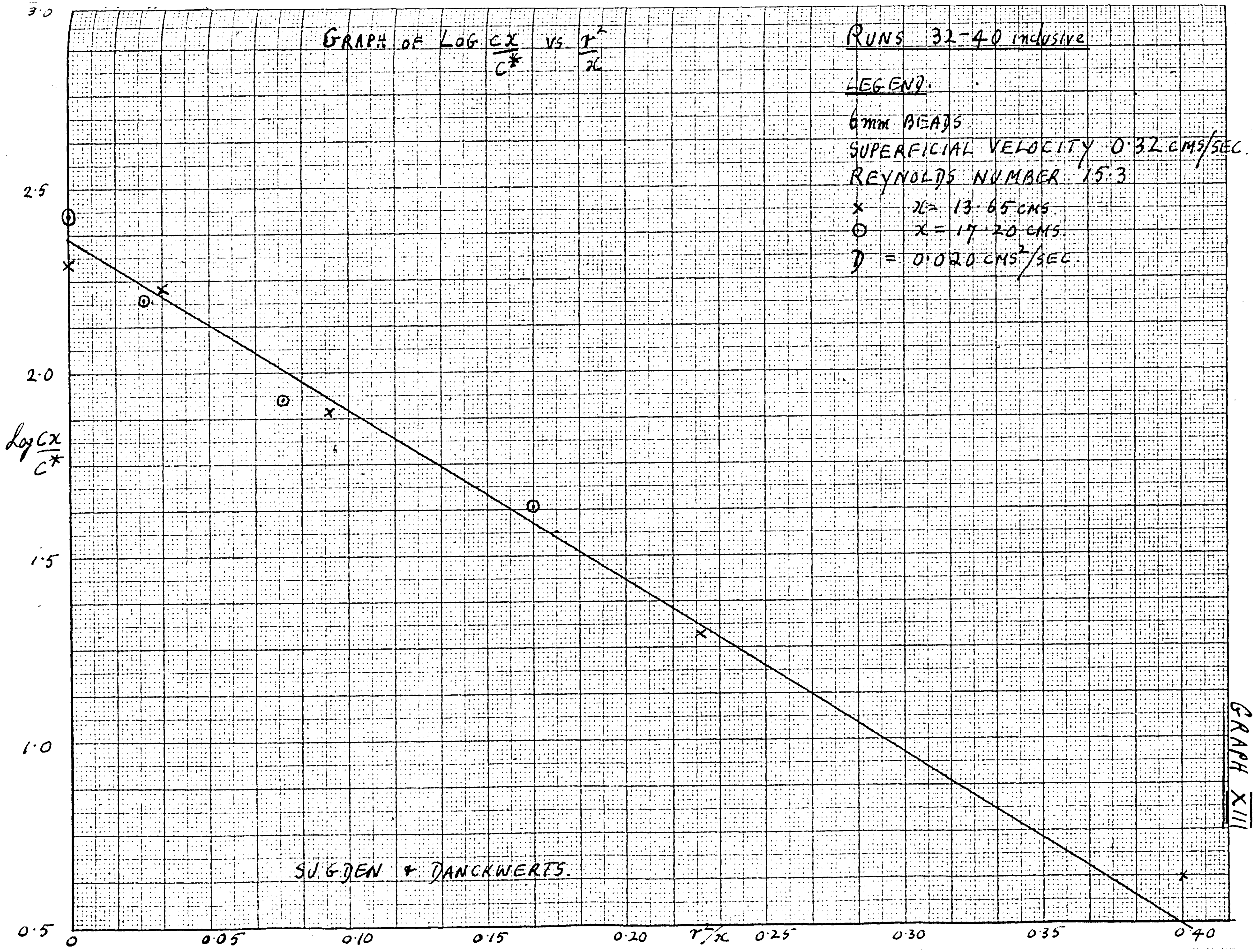
GRAPH XI


GRAPH OF $\log \frac{CX}{C^*}$ vs $\frac{r^2}{X}$

RUNS 16-23 inclusive

GRAPH XII


GRAPH OF $\log \frac{Cx}{C^*}$ vs $\frac{r^2}{x}$.
Runs 24-31 inclusive.
LEGEND:
6 mm. BEADS.
SUPERFICIAL VELOCITY 0·253 CM/SEC.
REYNOLDS NUMBER 11·99
⊙ x = 12·3 CMS
× x = 17·15 CMS.
D = 0·0103 CM²/SEC.
SUGDEN & DANCKWERTS
$\log \frac{Cx}{C^*}$
2·5
2·0
1·5
1·0
0·5
0
0·05
0·10
0·15
0·20
0·25
0·30
r^2/x
GRAPH XII

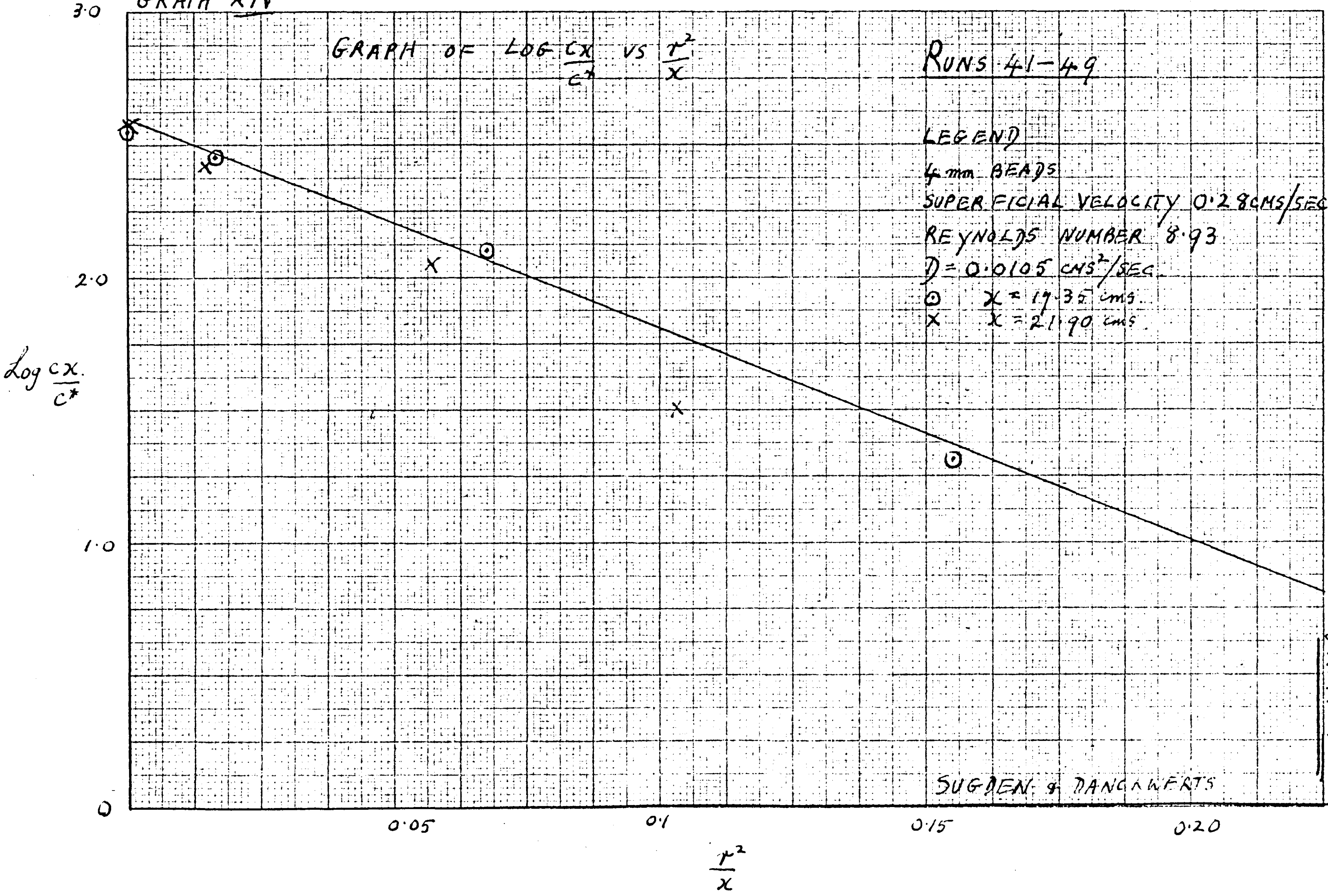


GRAPH OF LOG $\frac{cx}{c^*}$ vs $\frac{r^2}{x}$

RUNS 32-40 inclusive

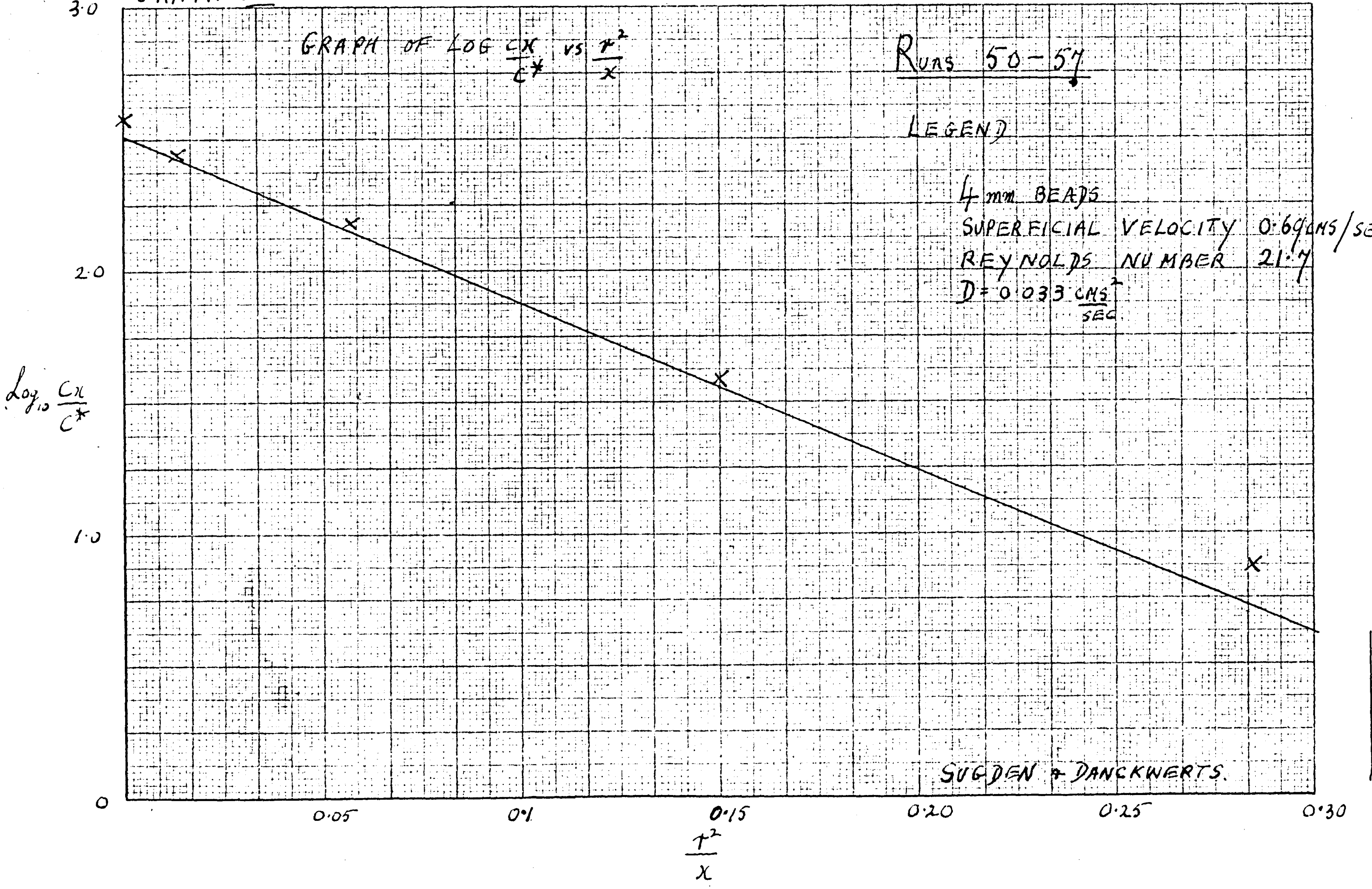
GRAPH XIII

GRAPH XIV



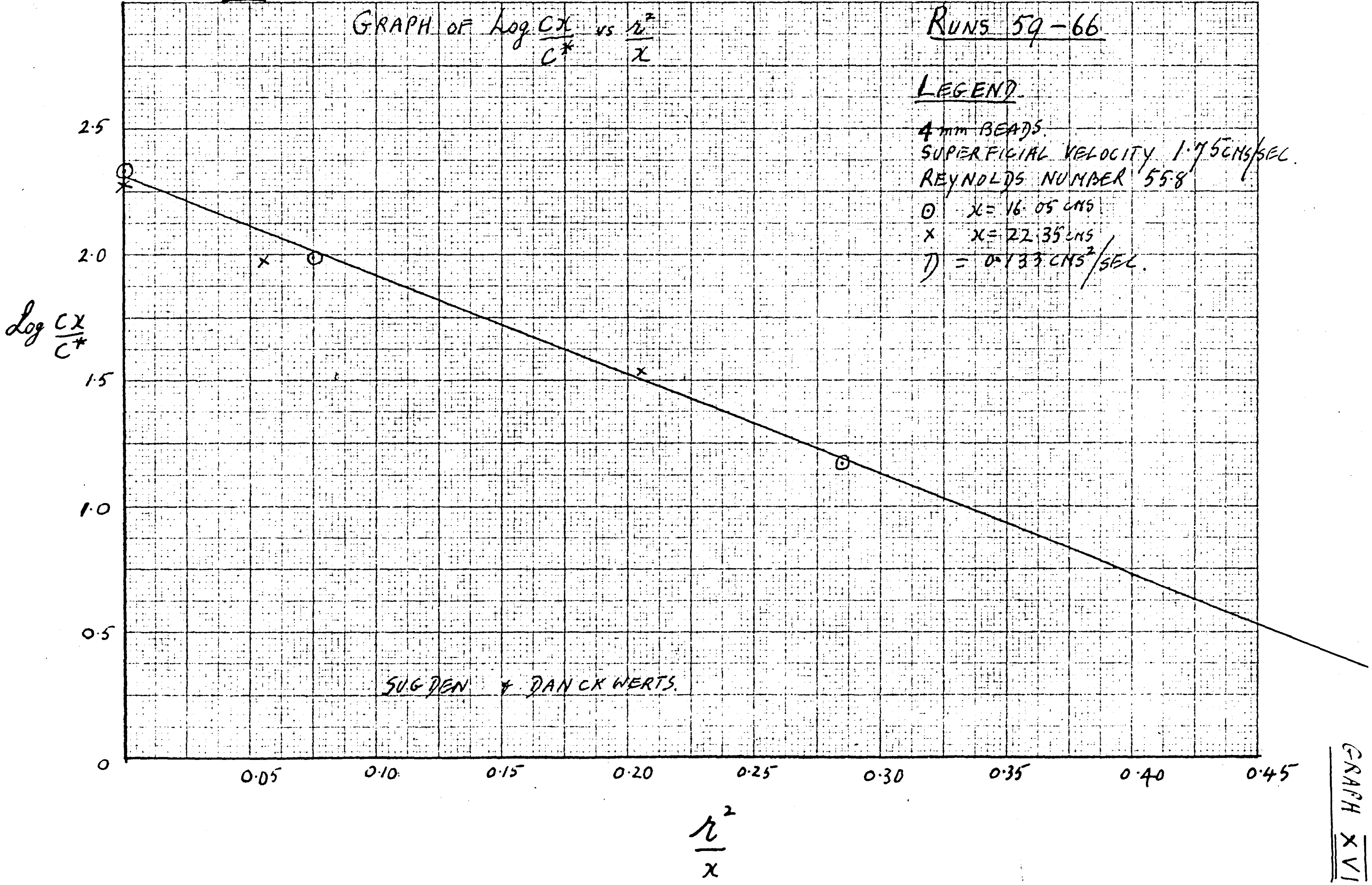
GRAPH XIV

GRAPH XV

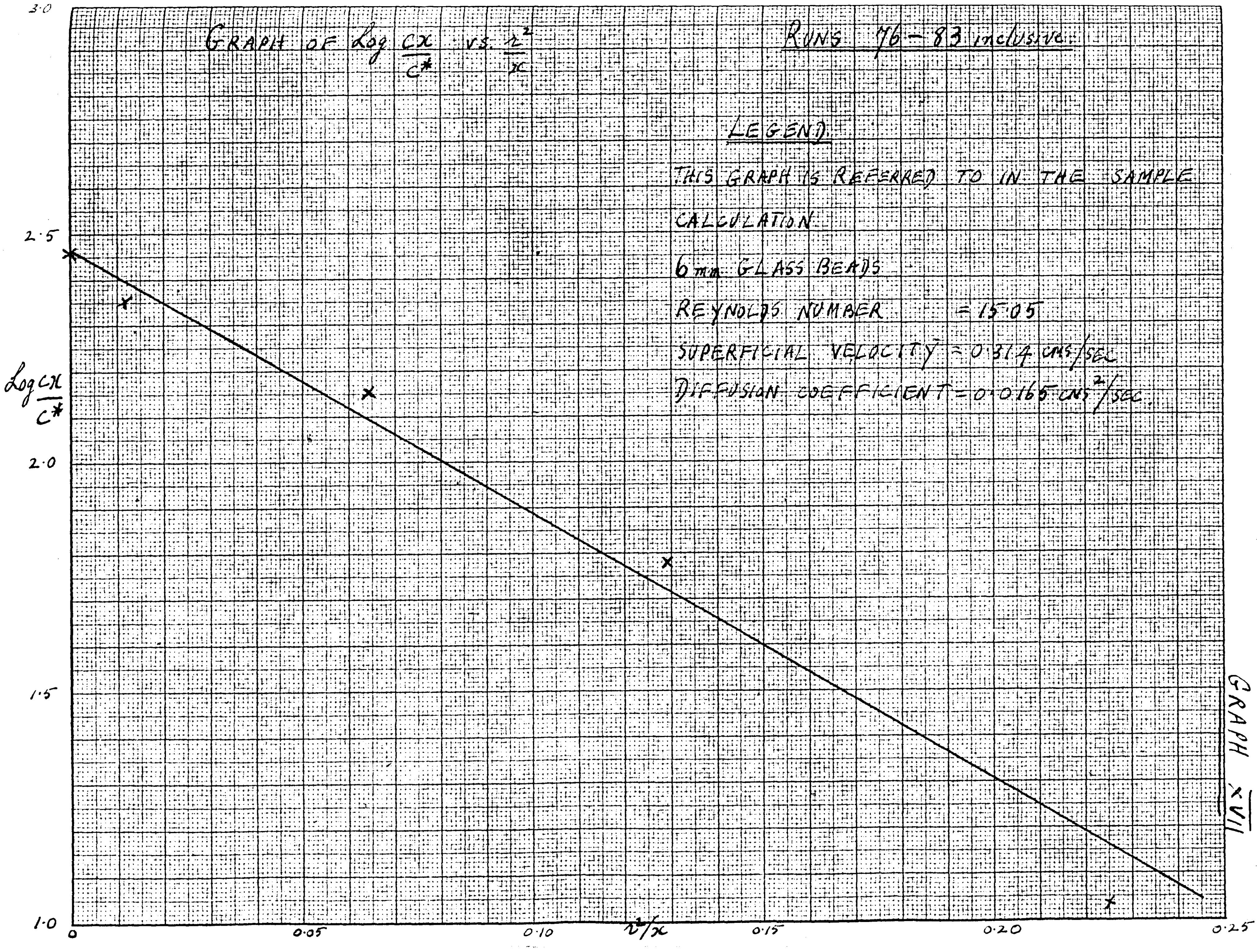


GRAPH XV

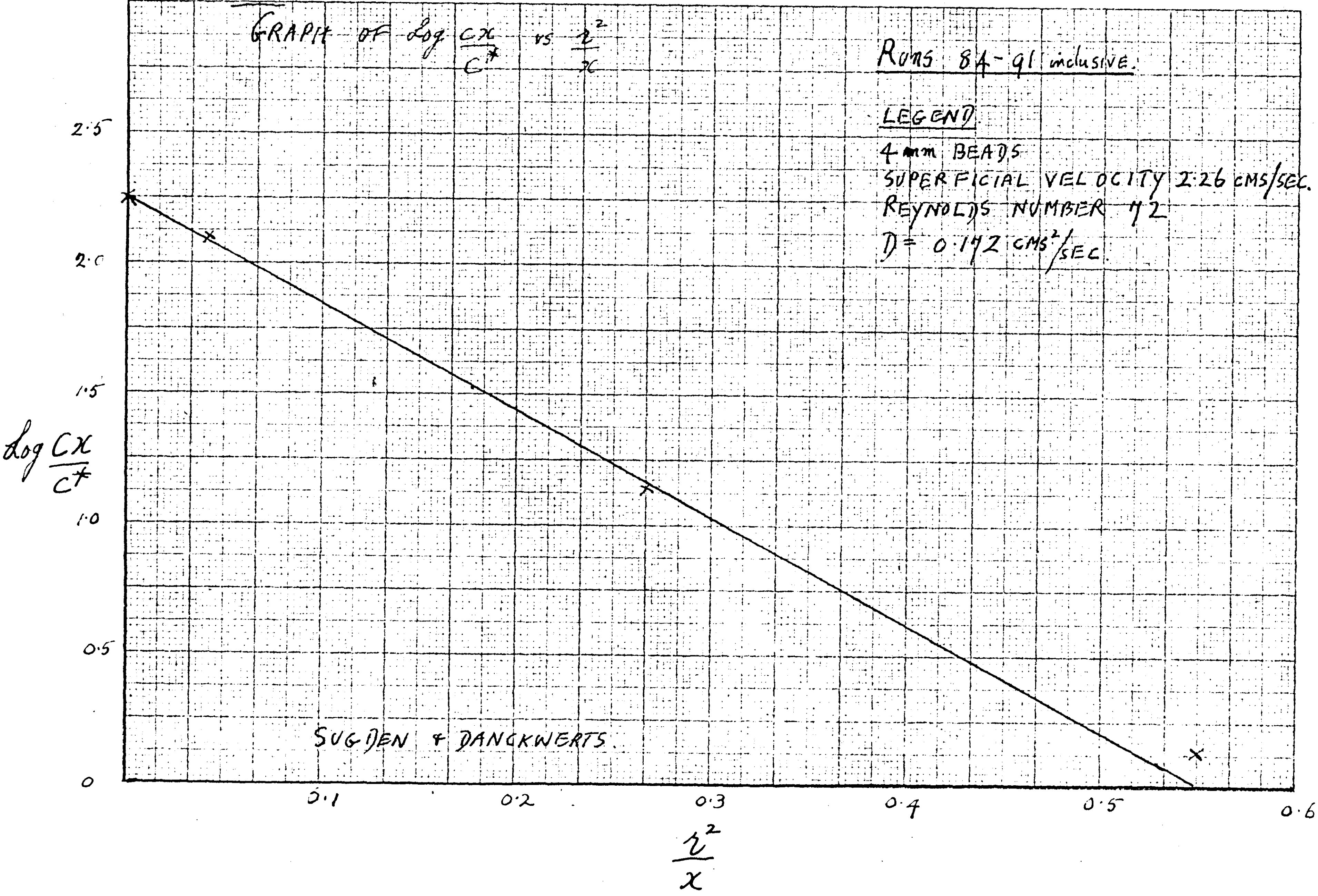
GRAPH XVI



GRAPH XVI

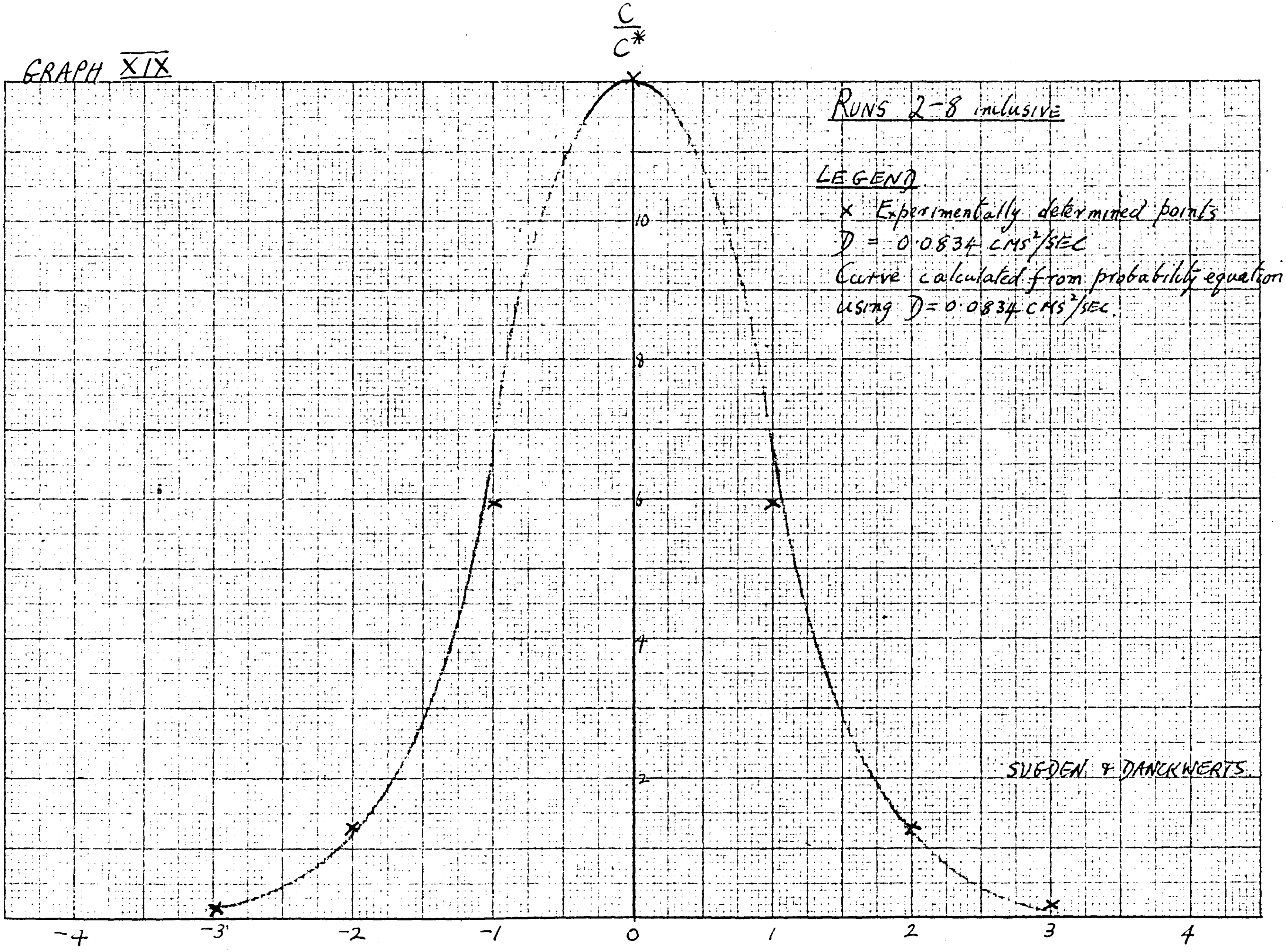


GRAPH XVIII



GRAPH XVIII

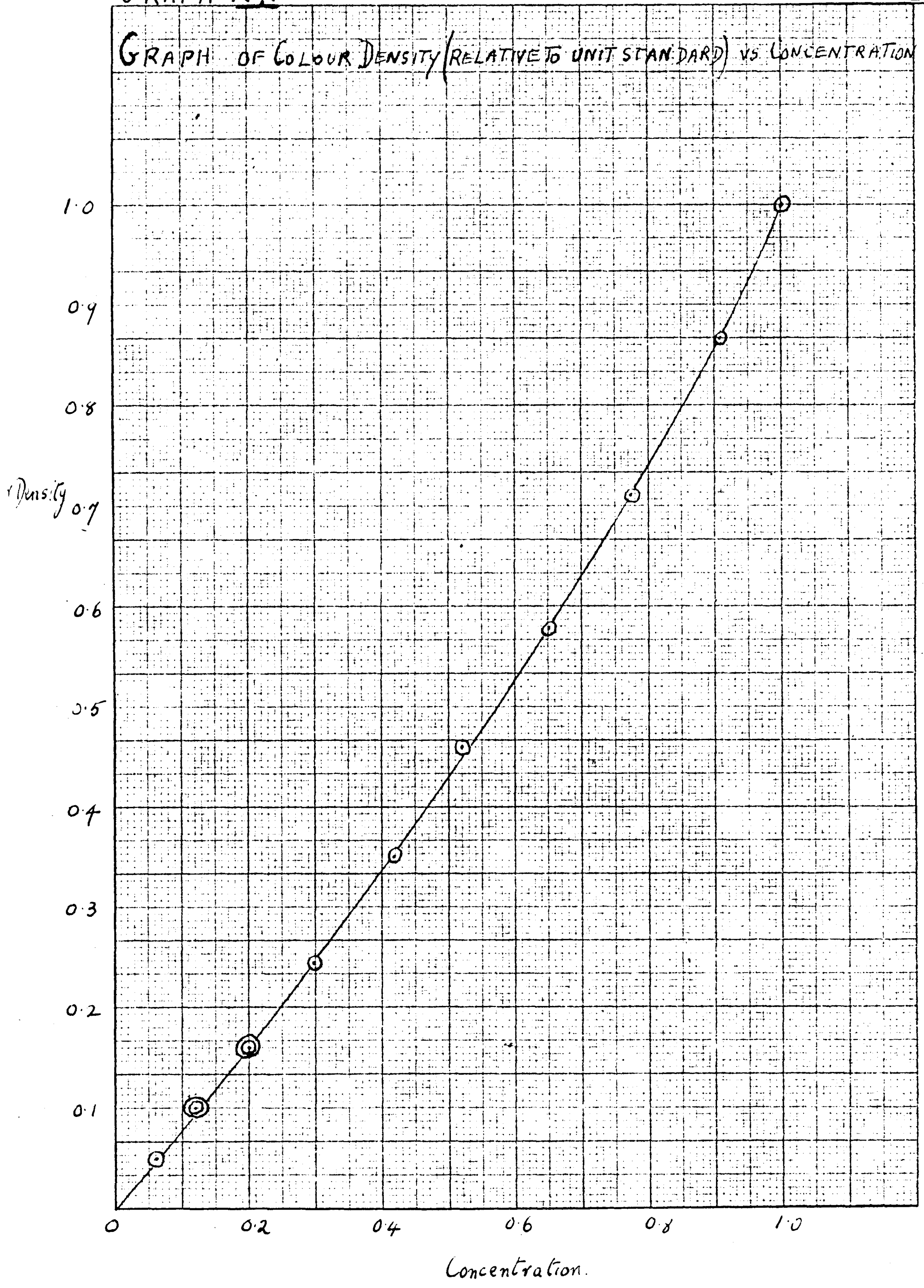
GRAPH XIX



GRAPH XIX

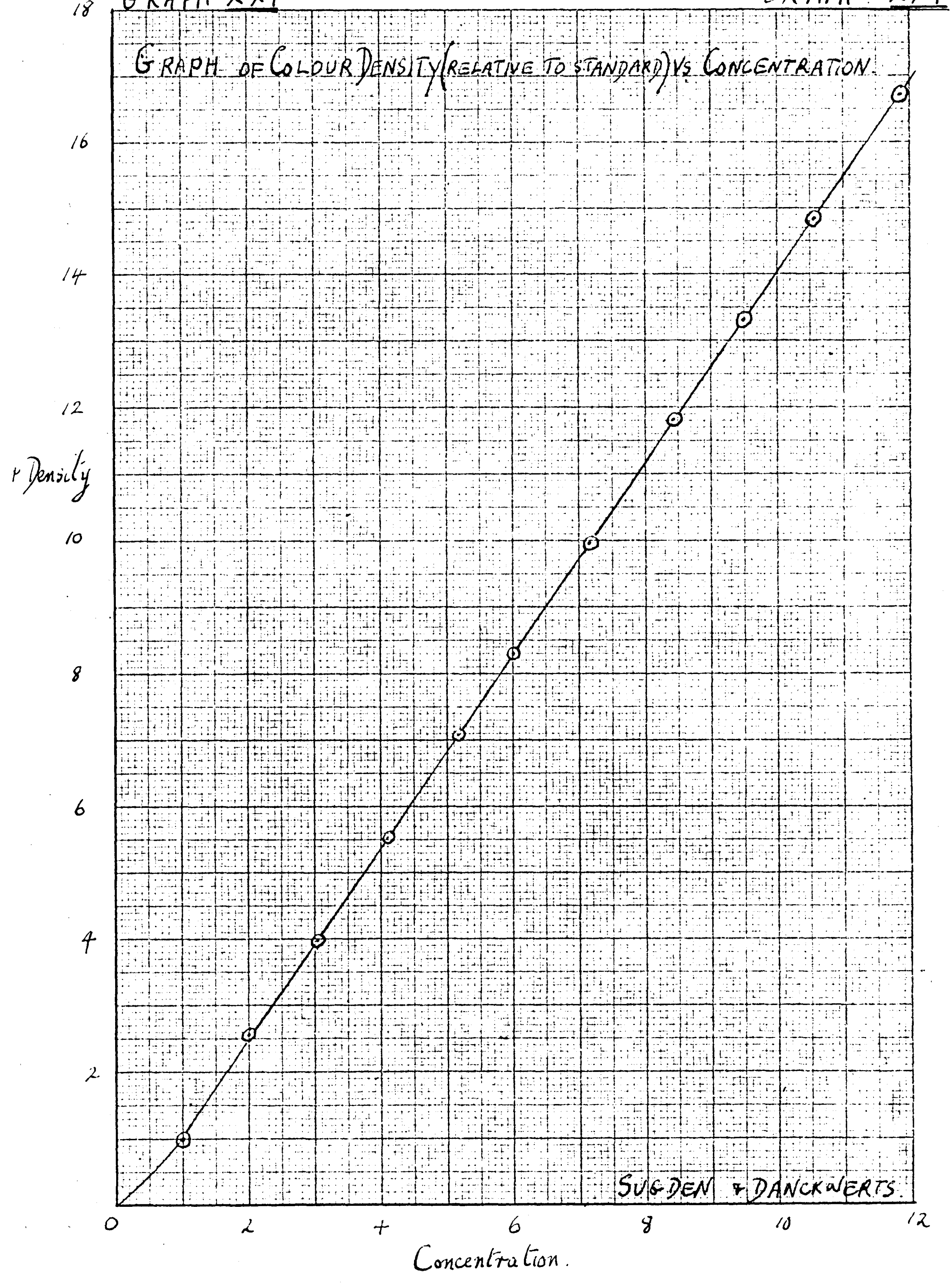
GRAPH XX

GRAPH XX

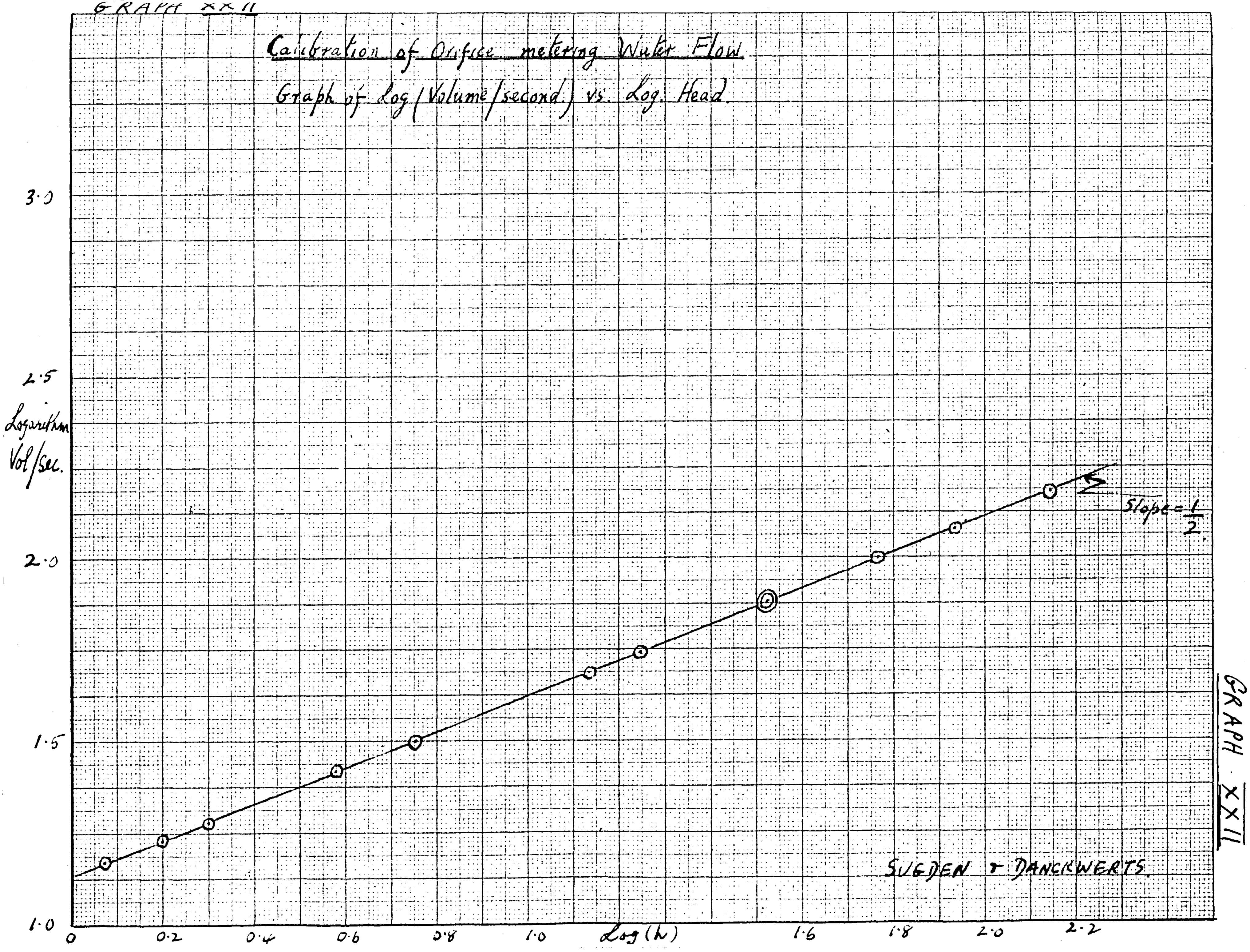

GRAPH OF COLOUR DENSITY (RELATIVE TO UNIT STANDARD) VS CONCENTRATION

GRAPH XXI

GRAPH XXI

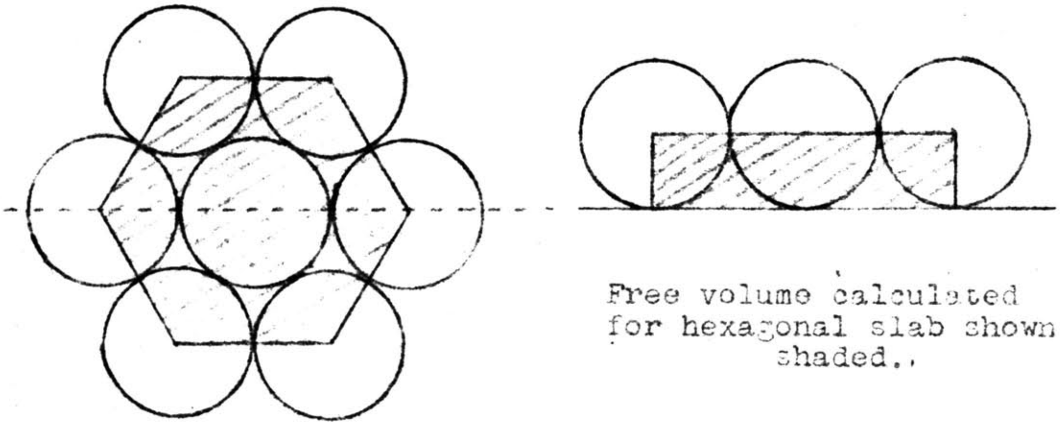


GRAPH XXII



Calibration of Orifice metering Water Flow

Graph of Log (Volume/second) vs. Log. Head



Free volume calculated for hexagonal slab shown shaded..

FIG: I

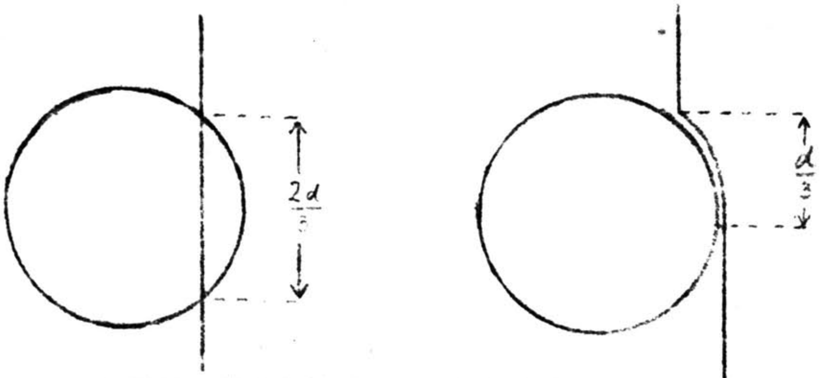


FIG: II Skirting and cutting paths.

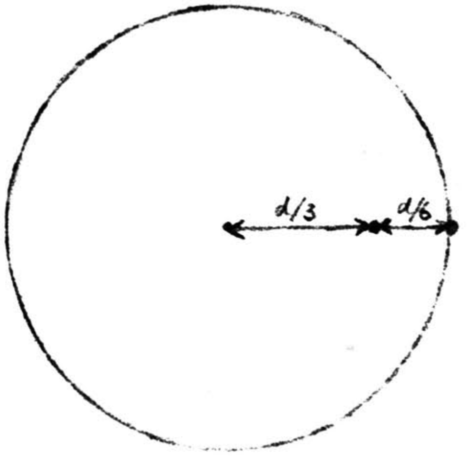


FIG: III Mean lateral displacement on great-circle path = d/6.

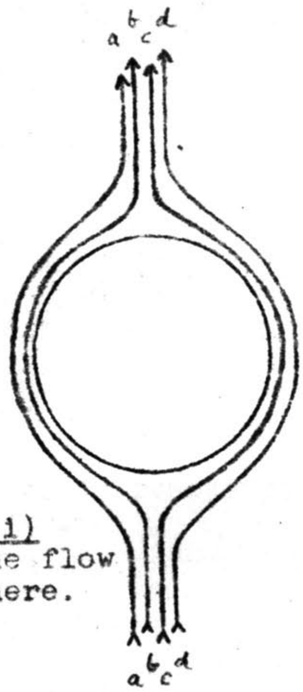


FIG: IV (i)
Streamline flow past sphere.

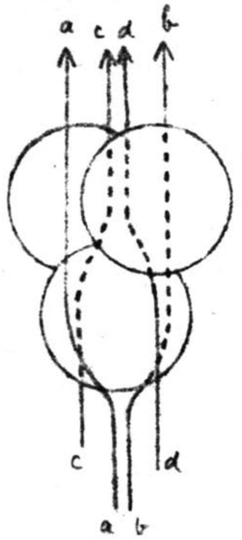


FIG: IV (ii)
Streamline flow in packing.

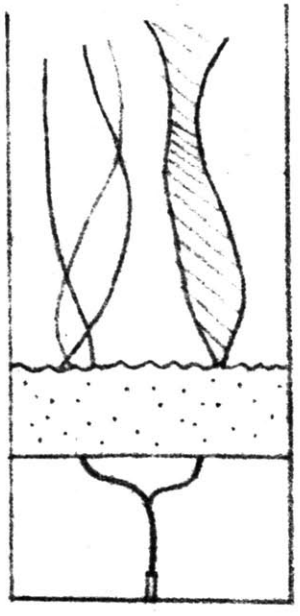
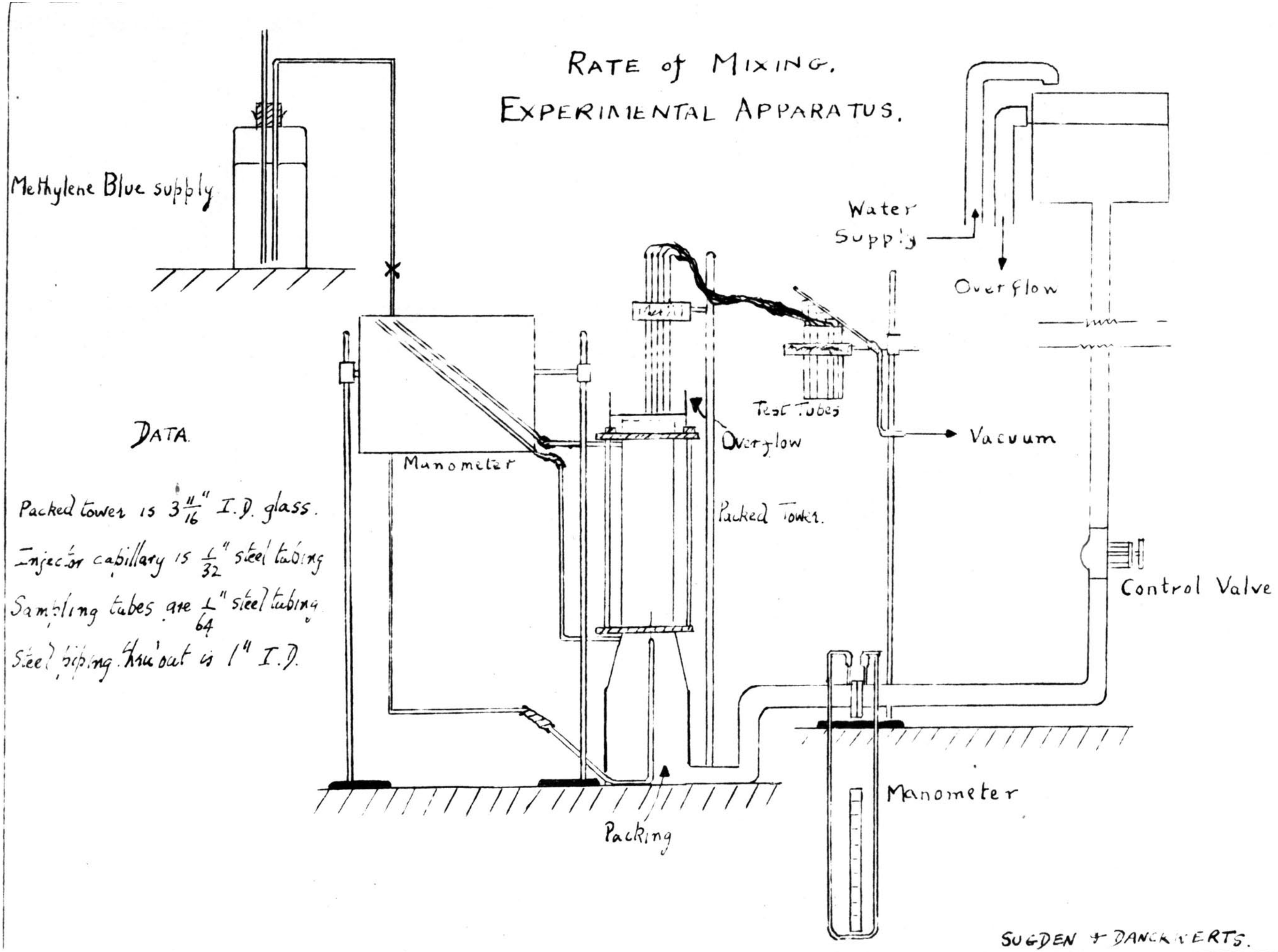


FIG: V.
Disturbance of flow by packing.

RATE of MIXING.
EXPERIMENTAL APPARATUS.



DATA.

Packed tower is $3\frac{11}{16}$" I.D. glass.

Injector capillary is $\frac{1}{32}$" steel tubing

Sampling tubes are $\frac{1}{64}$" steel tubing

Steel piping thru'out is 1" I.D.

FIG. VI

SUGDEN + DANCKWERTS.

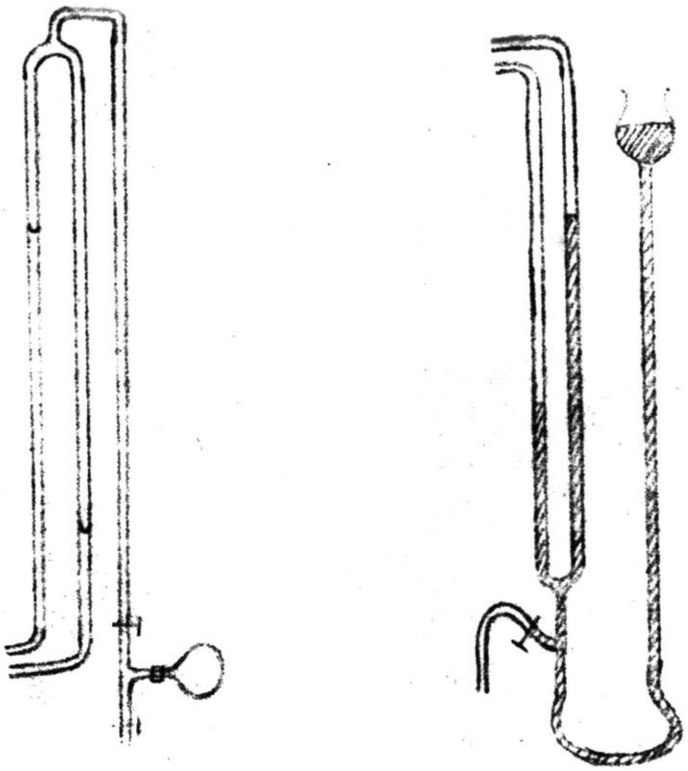


FIG: VII.
Water manometer. Mercury manometer.

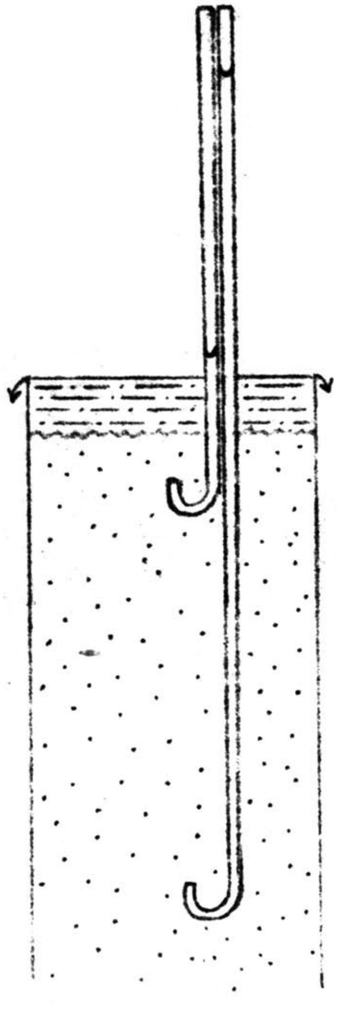


FIG: VIII.
Apparatus for measuring pressure-gradient through packing.

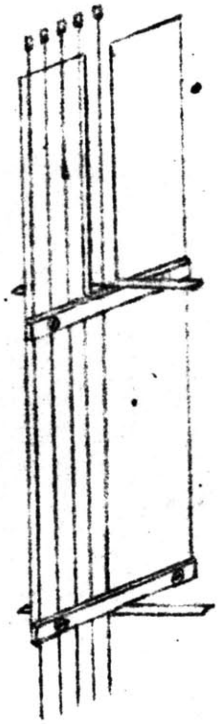


FIG: IX
Holder for sampling tubes.

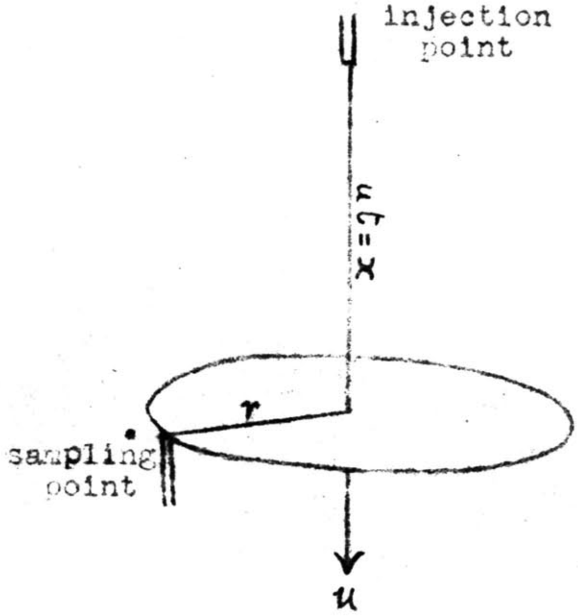


FIG: X

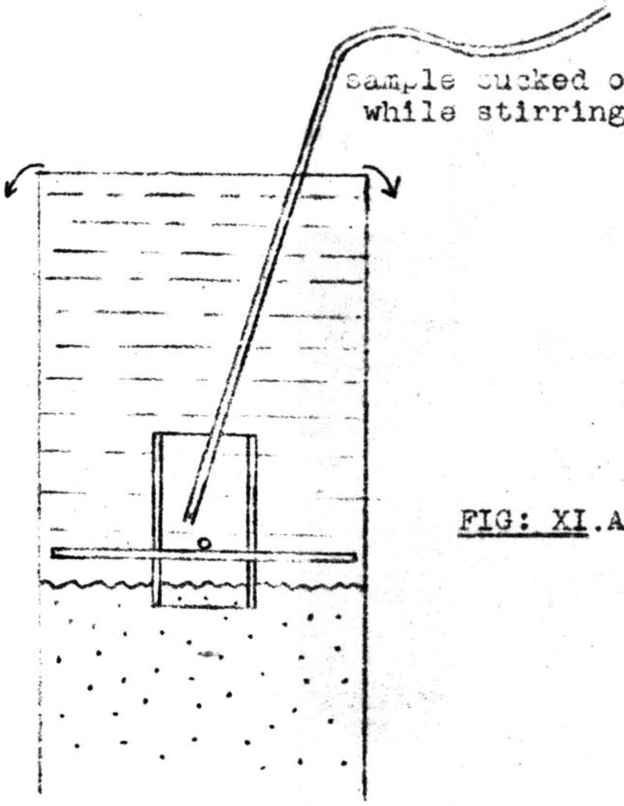


FIG: XI.Area sampling device.