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CATAPHORESIS OF LAMPBLACK

by

Jean Irwin Wagner

Submitted in Partial Fulfillment of the
Requirements for the Degree of

MASTER OF SCIENCE

from the
MASSACHUSETTS INSTITUTE OF TECHNOLOGY

1938

Signature of Author **Signature redacted**

Department of Chemical Engineering

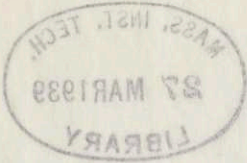
Signature of Professor **Signature redacted**

in Charge of Research

Signature of Chairman of Department
Committee on Graduate Students

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MR ✓



Chem. eng. Thesis
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Signature of Author

Jean Irwin Wagner

Department of Chemical Engineering

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Signature of Professor
in Charge of Research

F. B. ...

.....
Signature of Chairman of Department
Committee on Graduate Students

Harding Terrace
Centerport, New York
May 14, 1938.

Professor George W. Swett
Secretary of the Faculty
Massachusetts Institute of Technology
Cambridge, Massachusetts

Dear Sir:

I hereby submit a thesis entitled "Cataphoresis of Lampblack" in partial fulfillment of the requirements for the degree of Master of Science.

Respectfully yours,

Signature redacted

Jean Irwin Wagner

227711

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The author wishes to express his appreciation of the very helpful advice and suggestions which were given by Dr. Ernst A. Hauser under whom this investigation was carried out. He also wishes to thank the Dewey and Almy Chemical Company for furnishing the dispersing agent used in the experiments.

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I. SUMMARY

The following properties of aqueous solutions of the dispersing agent disodiumdinaphthylmethane disulphonate were measured; pH, surface tension, and viscosity. Dispersions of lampblack were prepared using these solutions. The relative amount of lampblack which was dispersed and the cataphoretic velocity of the suspended particles were determined. From the cataphoretic velocity, the zeta potential and the surface density of charge were calculated.

The results indicate that the dispersing agent behaves as though it were ionadsorbed on the dispersed particles which results in increasing the zeta potential and surface density of charge considerably above the values obtained in an ordinary electrolyte like sodium sulphate. The most important factor governing the dispersing action is the surface charge density to which the dispersing power is directly proportional in dilute solutions of the agent.

The results also suggest that perhaps the stability of the suspensoids is directly related to the density of charge.

II. INTRODUCTION

It is well known that properties of substances in the colloidal state show a distinct change as compared with their properties in other states of matter. For this reason many substances which are insoluble in water are used as finely divided aqueous suspensions, the particles of which average 0.5 to 3 microns in diameter and hence are in the upper limit of the colloidal range or just slightly above. The latter suspensions have been termed mechanical turbidities or cloudy systems and are characterized by an increased reactivity of the dispersed phase, by a maximum strength of color, covering power etc. Examples of particular applications of these systems are found in ceramics, pigments, water insoluble dyestuffs (e.g. vat and acetate silk dyestuffs), compounding ingredients for rubber latex processes, and horticultural insecticidal preparations. An example of the increased reactivity of a dispersed material is afforded by dispersed sulphur. If dried precipitated sulphur is mixed with some plain water, it may be kept in an ordinary clear glass container indefinitely without any sign of reaction. If a sample of the same sulphur is mixed with a small quantity of a dispersing agent and is ground in a suitable mill

with some water, the resulting dispersion will react with the small amount of iron in the glass and produce a gray-black deposit on the walls of the container.

The method most commonly used in the preparation of such dispersions is mechanical disintegration of the solid in the presence of water. This process is usually carried out in suitable grinding apparatus (e.g. ball mills, pebble mills, colloid mills etc.) and is known as wet grinding. If grinding is prolonged, it is observed that the reverse of the desired effect is obtained, namely that mechanical coagulation of the comminuted particles occurs, the result of which is a coarser suspension than is obtained by shorter grinding.

It has been found that the addition of certain water soluble substances tends to eliminate mechanical coagulation and to produce more stable dispersions. For example, lactose, glucose, and urea are known to aid the dispersion of indigo, sulphur, selenium, and aniline black (10). Recently, numerous long chain high molecular weight organic compounds have appeared under the term "dispersing agents." A. Stewart and H.M. Bunbury (9) state that these include the sulphonated oils, condensation products of aldehydes with aromatic sulphonates (e.g. disodium-

dinaphthylmethane-disulphonate which is prepared by condensing sulphonated naphthalene with formaldehyde), sulphated alcohols, sulphite cellulose lye and modified forms of this material. They also mention that soaps and proteins are often used but that these have certain disadvantages when compared with the synthetic products. S. G. Mark and W. F. Russell (6) limit the term dispersing agent to those substances which aid in the production of fine dispersions and which are not capillary active to any great extent. This is in disagreement with Stewart and Bunbury who include typical surface-active substances such as the sulphated alcohols and the sulphonated oils in their list of dispersing agents. It would seem logical therefore, to classify dispersing agents as surface-active and non surface-active. Examples of surface active dispersing agents have been given above, while disodiumdinaphthylmethane-disulphonate and salts of lignin sulphonic acid are examples of dispersing agents with practically negligible surface activity.

In addition to increasing the degree of comminution of the dispersed phase, these agents have other marked effects on the physical properties of suspensions. For example, if sulphur is ground with enough plain water to make a paste, the resulting product will have a coarse, flocculated appearance and

if it is diluted with water the particles will form flocs of a coarse net-like structure which will settle out very rapidly; but if a dispersing agent is used with the same amount of sulphur and water, the paste will have a smooth, even appearance and when diluted, the particles will not flocculate but will slowly settle out to form a clay-like deposit. It will also be noticed that the paste made with the dispersing agent is more fluid than the one prepared without it. In other words, it is possible through the use of dispersing agents to make a paste of a given fluidity or consistency using less water than one made without the addition of such compounds. It also follows that the maximum amount of a solid which can be dispersed in a given quantity of water is increased by the use of a dispersing agent.

As would be expected, the addition of electrolytes causes the flocculation of substances dispersed with these agents. The amounts needed to do this, however, are remarkably large.

A. Stewart and H. M. Bunbury (9) state that a dye-stuff suspension in a 1/1000 molal solution of dinaphthylmethane-disulphonate required a 1/125

molal concentration of sodium chloride just to produce flocculation, and in a 1/500 molal solution of the agent, the concentration of salt needed was 1/50 molal. This would indicate that the agent has a definite protective effect which would explain to a certain extent the mechanism of the dispersing action. They also state that dispersing agents probably owe their effects to being strongly adsorbed at the particle surface with their hydrophilic polar groups oriented toward the water phase. The hydrophobic nature of the surface is thereby reduced with a corresponding decrease in van der Waals forces and electrostatic effects which cause flocculation. It is also their opinion that these agents probably also increase the charge on the particles which is normally negative and when present in sufficient amount cause deflocculation.

J. G. Mark and W. F. Russell (6) are of the same opinion, stating that the agents probably put sufficiently large, like charges on the individual particles so that these repel each other and thereby overcome the cohesive forces between primary particles, thus breaking up flocs and forcing each particle to

act as a separate entity. The result is a true dispersion containing particles of optimum fineness with no appreciable aggregation remaining in the system. J. Reilly and W. N. Rae (7) state that the purpose of dispersing agents is, by physical or chemical means, to aid or accelerate the disintegration of the particles being subjected to a mechanical dispersing action. It is their opinion that theoretically the agents do this by precipitating or by combining with the electrolytes which are present in the material being disintegrated and which tend to cause flocculation of the comminuted material. They also state that actually the agents appear to act as protective colloids.

Since no experimental evidence was offered in support of the above theories, it was decided that this thesis should be a study of the deflocculating power of a dispersing agent and its relation to the charge on the dispersed particles and to other properties such as surface tension, and pH of aqueous solutions of the agent. Disodiumdinaphthylmethane-disulphonate was selected as the dispersing agent to be used because it is commercially available and is known to disperse lampblack (6).

The deflocculating or dispersing power of

the agent was determined by an application of Stokes' law of sedimentation which is expressed by the equation

$$V = \frac{2}{9} gr^2 \left(\frac{d_p - d_l}{\eta} \right)$$

where V = velocity of sedimentation, r = radius of the settling particle, d_p = density of the particle, d_l = density of the suspending liquid, g = gravity constant and η = viscosity of the liquid. From this, it follows that if a heterogeneous dispersion of particles of a given material in a given liquid is allowed to settle under the influence of gravity, the particles will settle out with a velocity proportional to the square of their radius. In order to obtain the relative fineness of dispersions with the same concentration of dispersed phase it is only necessary to allow equal volumes of each to stand in vessels of equal dimensions for a definite length of time and at the end of that time to determine the concentration of the solid at a definite distance from the surface of each dispersion. The concentration is proportional to the number of particles of equal radius which have settled to the level at which the concentration is determined.

The zeta potential of a suspended spherical particle has been expressed by D. C. Henry (4) in the form of the equation

$$\zeta = \frac{v6\pi\eta(2\Lambda_L + \lambda)}{D \cdot 3\Lambda_L}$$

where ζ = zeta potential (in e.s.u.), v = cataphoretic velocity of the particle per unit potential gradient, η = coefficient of viscosity of the dispersing medium, D = dielectric constant of the dispersing medium, Λ_L = specific conductance of the dispersing medium and λ = specific conductance of the suspended particle. Assuming that the suspended particles have a specific conductance very nearly equal to zero, the expression now becomes the familiar

$$\zeta = \frac{4\pi\eta v}{D} \quad (1)$$

H. A. Abramson (2) has shown that for large particles in dilute electrolytes the surface density of charge on the particles may be calculated from the equation

$$\sigma = \sqrt{\frac{NDkT}{2\pi \times 1000} \cdot c_{1[2]} z_{1[2]} \left[\frac{1}{z_2} \left(e^{z_2 \frac{E\zeta}{kT}} - 1 \right) + \frac{1}{z_1} \left(e^{-z_1 \frac{E\zeta}{kT}} - 1 \right) \right]}$$

where σ = surface density of charge in e.s.u./cm.²

N = Avogadro's number

- D = dielectric constant of the dispersing medium
- k = Boltzmanns constant
- T = absolute temperature
- $c_1(2)$ = molal concentration of ions
- $z_1(2)$ = valence of the ions
- E = elementary charge
- ζ = zeta potential

For a uni-divalent electrolyte at 25°C, $z_2 = 2$
and the equation reduces to

$$\sigma = 17900 \sqrt{c_2 \cdot \sqrt{e^{77.6 \zeta} + 2e^{-38.8 \zeta} - 3}} \quad (2.)$$

H. A. Abramson (3) has also shown that the course of the $\sigma - c$ curve in dilute solutions of simple electrolytes very closely follows a curve similar to the Langmuir adsorption isotherm and which may be represented by the equation

$$\sigma = \sigma_M \frac{\beta' c}{1 + \beta' c} \quad (3)$$

where σ_M = the limiting value of σ

$\sigma_M \beta'$ = the slope of the $\sigma - c$ curve in dilute solution,

and β' = a constant

III. PROCEDURE

Solutions of the dispersing agent were prepared as follows: 200 grams of commercial disodium - dinaphthylmethane-disulphonate (Daxad 11, Dewey and Almy Chemical Co.) were dissolved in 900 millilitres of hot distilled water and the solution filtered and evaporated to dryness. The material was then powdered and dried at 110° C for one hour.

The dispersing agent was analyzed for sulphate by the method of E. Hintz and H. Weber which is described by F. Treadwell and W. Hall (11). It was found to have 13.31% by weight of sulphate calculated as Na_2SO_4 . No chloride was found present.

Solutions were made with concentrations of the agent ranging from 0.0004 to 0.0922 molal (based on the formula weight of 472) by weighing the required amount of agent into a volumetric flask and adding distilled water to the mark.

In order to obtain the dispersing agent free from sodium sulphate, electro dialysis of a 30% solution was tried. It was found that the agent itself dialyzed as well as the electrolyte. The method used by C. Robinson and H.A.T. Mills (8) for the purification of benzopurpurin 2B was tried. This consisted of salting out the dye with sodium acetate and dissolving the acetate adhering to it with ethyl alcohol. In

this case, however, the dispersing agent would not salt out. It was finally found that the sulphate could be removed by precipitating it as barium sulphate with barium thiocyanate. The barium sulphate was centrifuged out in a tube type centrifuge and the solution was evaporated to dryness. The thiocyanates left in the agent were removed by extraction of the solid with ethyl alcohol in a Soxhlet extractor until no test for thiocyanate could be obtained by the addition of ferric chloride to a sample of the alcohol extract coming from the solid. The alcohol-wet solid dried at 105° C to a white powder. A water solution of it became slightly opalescent on the addition of barium chloride and 6N hydrochloric acid. Analysis for sulphate failed to give a weighable precipitate. Solutions were prepared from the dried solid ranging from 0.0005 to 0.100 molal in concentration. These solutions had a turbidity which was undoubtedly due to colloidal barium sulphate which had not settled out in centrifuging.

Suspensions of lampblack were prepared by shaking for one minute 0.100 grams of the solid with exactly 100 millilitres of each of the solutions of the dispersing agent. They were allowed to stand three days and at the end of that time the relative degree of dispersion was determined by a Lange photo-

electric colorimeter. Five millilitres of the solution of the dispersing agent used in the preparation of the suspension which was being measured was pipetted into each of the two glass cells and diluted to fifty millilitres. The zero reading was adjusted and the scale reading for an absorption of one hundred percent was set at eighty. The right-hand cell was then removed and cleaned. Five millilitres of the dispersion was withdrawn at a distance of one-half inch from the surface (this distance being kept constant for each determination) and diluted to fifty millilitres in the glass cell. The absorption was then read from the microammeter scale. This reading was converted to an absorption of one hundred percent by dividing by eight-tenths. The absorption value is a relative measure of the dispersing power of the agent at the particular concentration used.

The cataphoretic velocity of the suspended particles was measured in a glass cell of the Northrup-Kunitz type, as manufactured by A.H. Thomas Company, Philadelphia.

Calibration of the cell was carried out in the following manner. The zinc electrodes were filled with saturated zinc sulphate and the cell was

filled with one-tenth normal potassium chloride. Care was taken to exclude all air bubbles. A direct current voltmeter was connected across the zinc electrodes and a Lindemann electrometer across the sealed in platinum electrodes of the cell. It is necessary to use an electrometer of this type or one with equally high resistance in order to avoid polarization of the platinum electrodes. A potential was then applied to the zinc electrodes, the voltmeter reading recorded and the voltage drop through the cell determined with the Lindemann electrometer. Ten such determinations were made with applied voltages ranging from 9.90 to 17.80. The ratio of the drop in the cell to the applied voltage was calculated for each one and the arithmetic mean of them all was taken. This was found to be 0.22. For any determination, the potential gradient in the cell may be obtained by multiplying the applied voltage by this ratio and dividing by the distance between the platinum electrodes which was 35 mm. in this case.

The depth of the cell was obtained by direct measurement with a microscope micrometer. Four determinations were made at each end of the cell and in the middle. The arithmetic mean of each set was taken and the three figures obtained were averaged in a similar manner. This showed the

average depth of the cell to be 0.6460 mm. The width of the cell was 12mm. Substitution of these values in the equation of Komagata (1) gave the distance of the stationary layer from the top or bottom of the cell as 0.204 multiplied by the depth of the cell.

To measure the cataphoretic velocity of the carbon particles, the zinc electrode vessels were filled with saturated zinc sulphate solution, the cell was thoroughly washed with distilled water, then with 10ml. of the carbon suspension and finally was filled with the suspension. A Zeiss microscope fitted with a 40x water immersion objective and a 15x binocular eyepiece containing an eyepiece micrometer was focussed at the depth of the stationary layer. A potential was applied to the zinc electrodes and the time was taken for a particle to move a distance on the micrometer eyepiece scale equivalent to 45 microns in the cell. The current was reversed and the same procedure was again followed. For each suspension ten such determinations were made and the arithmetic mean of them all was taken. Only particles with diameters ranging approximately from 2 to 5 microns were selected for observation in each case. Large particles were selected for observation because

of better visability and because particle size does not enter into the equation for cataphoretic velocity.

The viscosity of solutions of the dispersing agent was measured with an Ostwald viscosimeter at $25^{\circ} \pm 0.1^{\circ}\text{C}$. The viscosity of the solutions of sodium sulphate was obtained from the International Critical Tables (5).

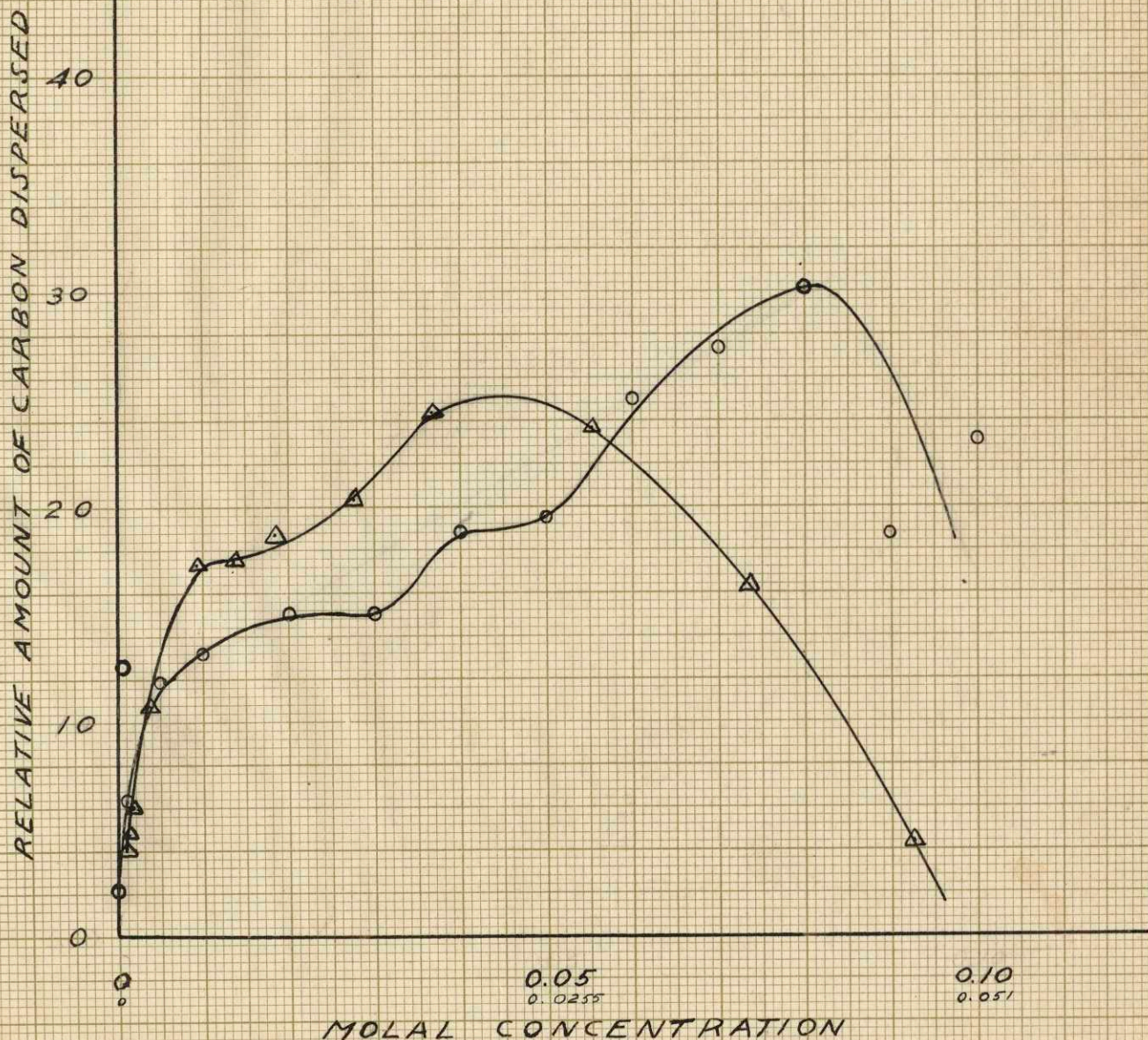
The hydrogen ion concentration in each of the solutions and in each of the dispersions made with them was measured with a Beckmann pH meter.

A du Nouy interfacial tensiometer was used to determine the surface tension of solutions of the salt-free dispersing agent. Two sets of data were taken. In one, the surface tension was measured at intervals of about one minute from the time immediately after pouring the solution into the vessel until constant readings were obtained. These data have been plotted as "readings taken to constant value" in Figure 5. In the other, readings were taken at definite time intervals up to 31 hours after pouring into the vessel. At each interval the first reading only was taken as the equilibrium value for that period.

Fig. 1

DISPERSING POWER

○ - of Dinaphthylmethanedisulphonate - free from Na_2SO_4
△ - " " " " " " + 13% Na_2SO_4 (by wgt)
The small numbers are the concentration of Na_2SO_4 in △



IV. RESULTS

The results are presented in the form of graphs in Figures 1 to 7. In each case the abscissa represents the molal concentration of the dispersing agent or electrolyte present in the solution or dispersion. Where a mixture of sodium sulphate and dispersing agent was used the molal concentration of the agent alone is given with the corresponding concentration of the electrolyte below it in small numbers. The ordinate represents the property or characteristic which was determined.

Figure 1 shows the effect of concentration of the dispersing agent on the dispersing power. It will be noticed that in very dilute solutions of the agent plus sodium sulphate the dispersing power increases very rapidly with concentration to 0.01 molal where it begins to increase less rapidly until a maximum is reached at 0.045 molal after which it falls rapidly. In the salt free dispersing agent a similar result is observed with the maximum coming 0.08 molal. In low concentrations the dispersing power of the salt free agent is less than that of the agent plus sodium sulphate, but as the concentration increases, it increases until a maximum is reached which is greater than the maximum of the agent plus sodium sulphate. It will also be noticed that up

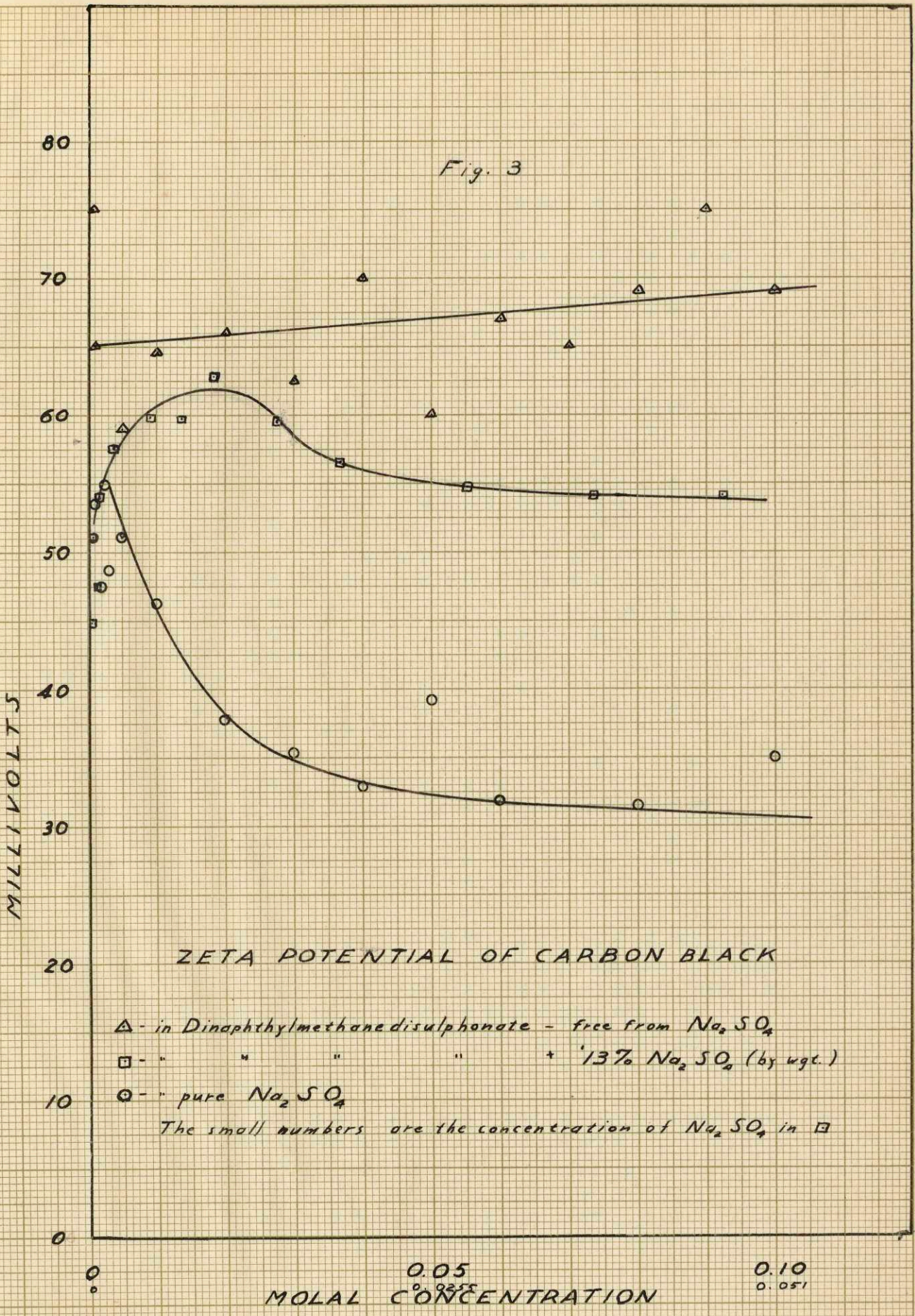
Fig. 2

pH OF DINAPHTHYLMETHANE DISULPHONATE SOLUTIONS
AND OF CARBON DISPERSIONS MADE WITH THEM

- Δ = Dinaphthylmethanedisulphonate + 13% Na_2SO_4
- X = Carbon dispersion made with Δ
- \square = Dinaphthylmethanedisulphonate - free from Na_2SO_4
- O = Carbon dispersion made with \square



Fig. 3



to 0.04 molal both of the curves resemble adsorption isotherms.

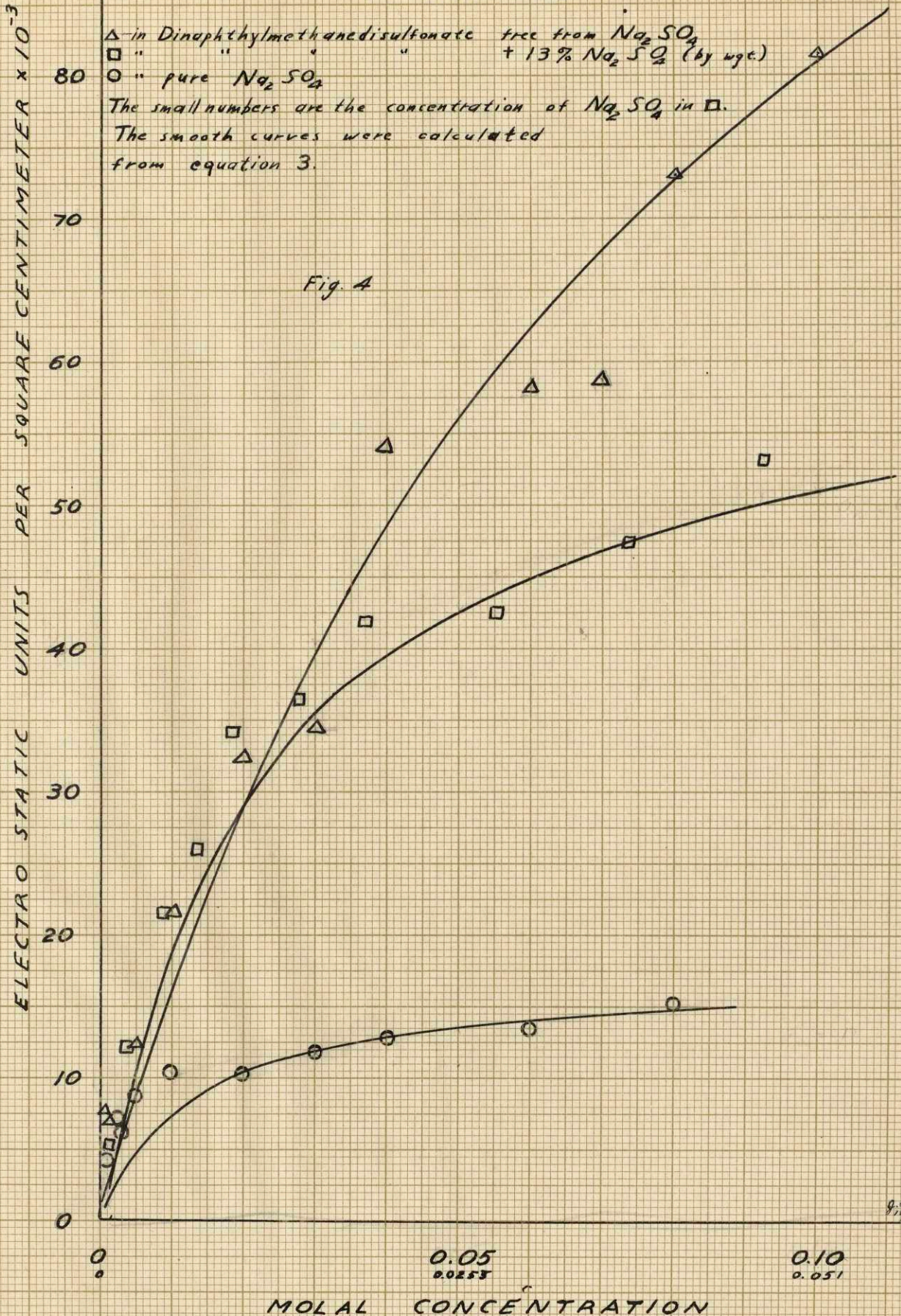
Figure 2 shows the variation of pH with concentration of solutions of the dispersing agent and of carbon dispersions made with them. The curves obtained from the agent plus sodium sulphate show a maximum at 0.045 molal, but in general the pH does not change much. The pH of the dispersions is very nearly identical with that of the solutions from which they were made. The values obtained in the salt free agent are considerably lower than in the agent plus sodium sulphate, and in general do not show any great variation from the dispersions made therewith.

The effect of the salt free dispersing agent, of a mixture of dispersing agent plus sodium sulphate and of pure sodium sulphate on the zeta potential of the carbon particles is shown in Figure 3. The zeta potential in pure sodium sulphate rises to a maximum of 55 millivolts at 0.003 molal then falls off, rapidly at first, then more slowly, and approaches a limiting potential of approximately 30 millivolts. In the dispersing agent plus sodium sulphate the zeta potential rises to a maximum of 63 millivolts at concentration of the agent of 0.019 molal, then falls off slowly and approaches a limiting potential of 53

SURFACE DENSITY OF CHARGE ON CARBON PARTICLES

Δ - in Dinaphthylmethanedisulfonate free from Na_2SO_4
 \square " " " " " " " " + 13% Na_2SO_4 (by wgt.)
 \circ " pure Na_2SO_4

The small numbers are the concentration of Na_2SO_4 in \square .
 The smooth curves were calculated from equation 3.



G. W. Stewart
9/12/32

millivolts. Both curves are typical for inert surfaces in simple electrolytes. There is little variation in potential in the case of the salt free agent, the curve being a straight line sloping slightly upward. At all concentrations the potential in the salt free agent is higher than in the agent plus sodium sulphate. The potential in pure sodium sulphate solutions is considerably lower than in the solutions of the dispersing agent plus sodium sulphate.

The surface density of charge on carbon particles in the same dispersions has been plotted in Figure 4. The plotted points were calculated from equation (2); the smooth curves from equation (3). In low concentrations the surface charge density is greater in solutions of the dispersing agent plus sodium sulphate than in the solutions of the salt free dispersing agent. At 0.02 molal the curves cross and the reverse is true. The surface charge density is considerably less in pure sodium sulphate than in the solutions of the salt free dispersing agent and in the dispersing agent plus sodium sulphate. It will be noticed that in general all three curves take the form of the adsorption isotherms calculated from equation (3).

Fig. 5
SURFACE TENSION OF SALT FREE
DINAPHTHYLMETHANE DISULPHONATE

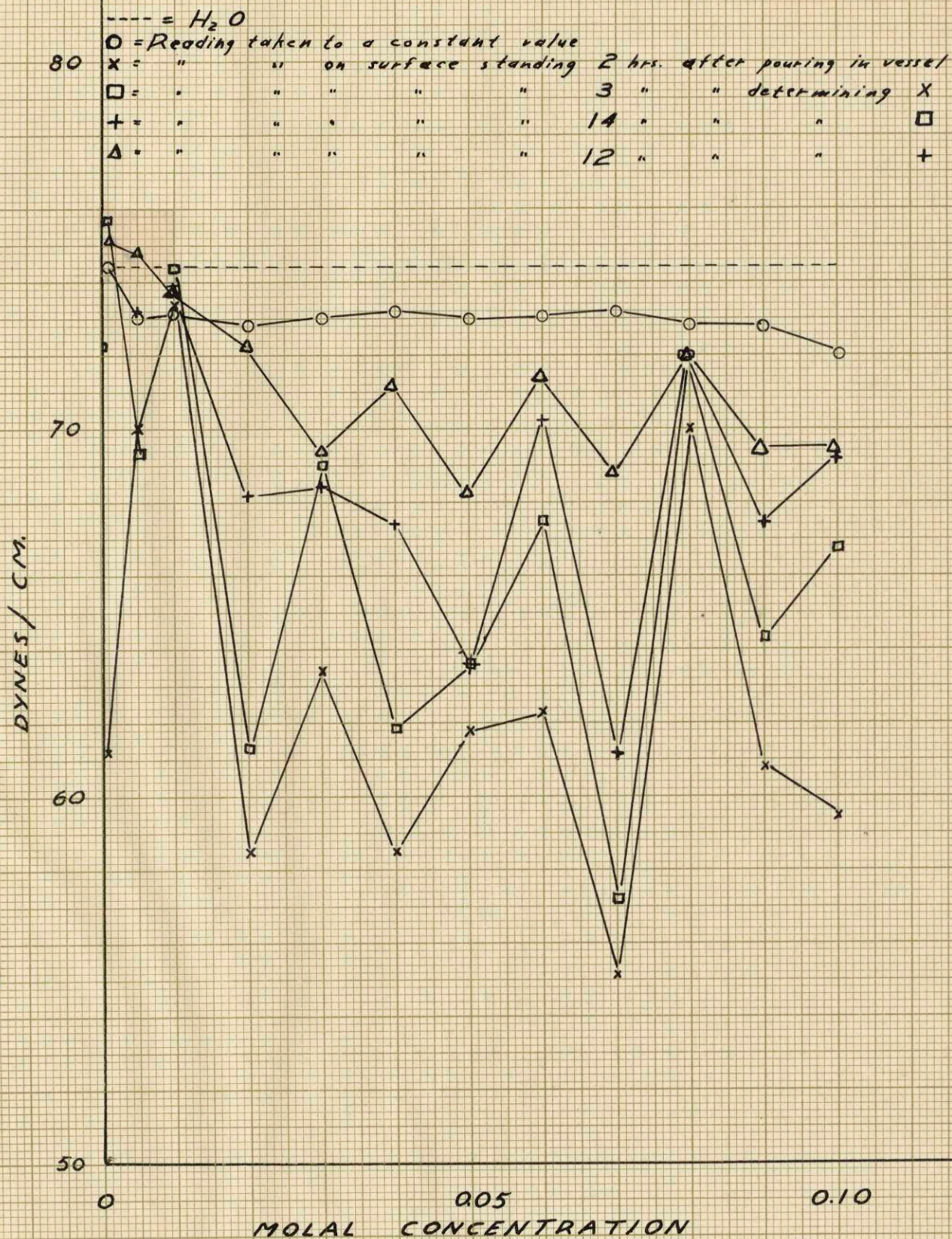
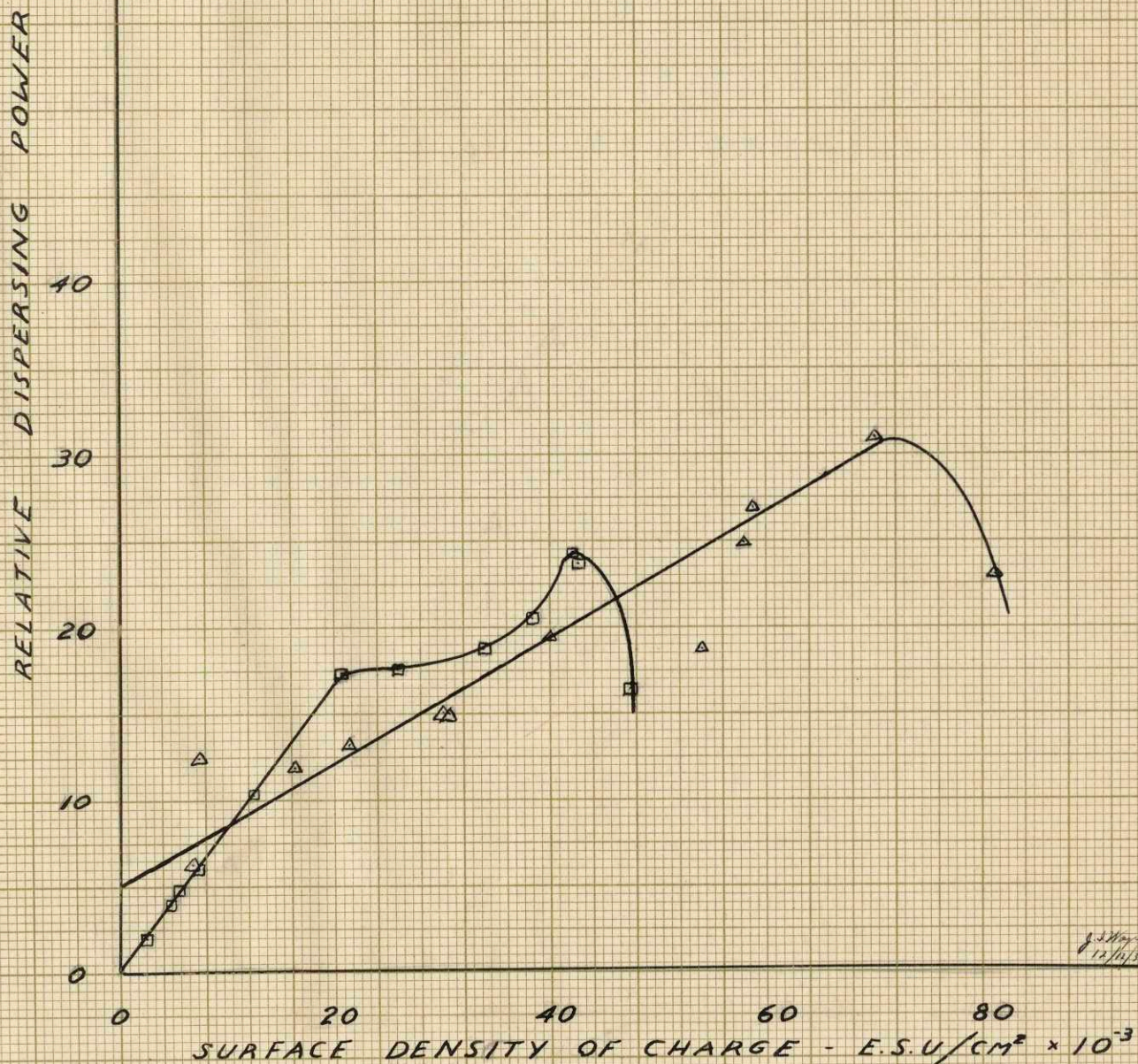


Figure 5 shows the variation of surface tension of solutions of the salt free agent with concentration and time. The readings taken to a constant value show uniform values of 73 ± 0.2 dynes/cm. at concentrations ranging from 0.005 to 0.06 molal. After the latter concentration, the surface tension begins to fall until at 0.10 molal it reaches 72 dynes/cm. The values taken at definite lengths of time after pouring show that after standing for two hours there is a marked reduction in surface tension which varies considerably with concentration. At 0.01 - 0.02 molal and at 0.07 - 0.08 molal the surface tension drops from 73.5 to 58.5 and from 70.0 to 55.0 respectively. These are the most extreme variations which occur. As time goes on, the variation decreases and the curves become more cyclic in nature so that 31 hours after pouring into the vessel, the surface tension varies from 68.4 to 71.5 to 68.8 dynes/cm. at concentrations of 0.05, 0.06, and 0.07 molal respectively. This variation is very nearly constant for the concentrations ranging from 0.03 to 0.09. However in extremely dilute solutions, the surface tension rises to 75.0 dynes/cm. (0.6 greater than the value found for water) at a concentration of 0.001 molal, then falls rapidly to 69.4 at 0.03 molal. In general, it

Fig. 6

COMPARISON OF DISPERSING POWER
WITH SURFACE DENSITY OF CHARGE

Δ - in disodiumdinaphthylmethane disulphonate free from Na_2SO_4
 \square " " " " + 13% Na_2SO_4

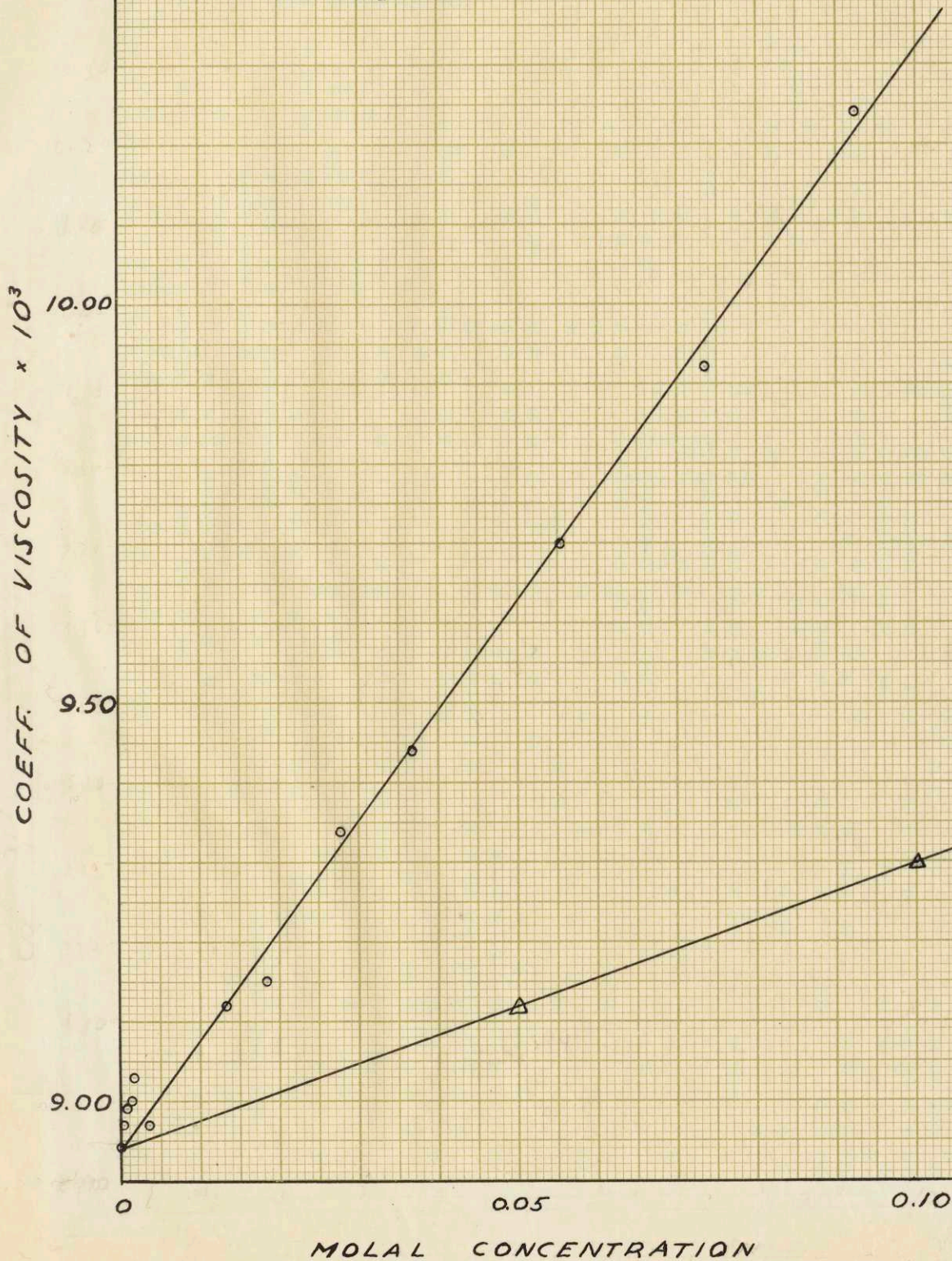


J. J. W. W.
12/14/52

Fig. 7

VISCOSITY AT 25°

○ - Dinaphthylmethane disulfonate + 13% Na₂SO₄
△ - Na₂SO₄



J. W. C. ...
4/11/33

will be noticed that after the initial reduction, the surface tension tends to approach the readings taken to a constant value.

In Figure 6, a comparison of the dispersing power with surface density of charge is made by plotting the relative dispersing power against the surface density of charge. In the case of the salt free dispersing agent, the dispersing power is directly proportional to the surface density of charge up to 0.08 molal after which concentration this relation does not hold. Similarly the dispersing power of the dispersing agent plus 13% sodium sulphate is directly proportional to the surface charge density up to 0.015 molal after which concentration this relation does not hold.

Figure 7 shows the variation of the viscosity of aqueous solutions of the dispersing agent with concentration of the agent in them. It will be observed that the curve is a straight line and that the viscosity of a 0.1 molal solution is approximately only 15% greater than that of water.

V. DISCUSSION OF RESULTS

In general the data obtained from the salt free dispersing agent do not follow as smooth curves as those from the agent plus sodium sulphate. This may be due to traces of barium sulphate which could not be removed by the centrifuging process used in the preparation of it. Reason to believe this is given by the fact that the carbon suspensions made with the salt free agent were a medium gray shade whereas those prepared with the agent plus sodium sulphate were black. This would indicate that the barium sulphate was adsorbed on the carbon particles and as a result it might affect their surface properties. There is also a possibility that some thiocyanate was not completely removed in the extraction with alcohol, but since the test for it is so sensitive, the possibility is small. The presence of enough alcohol to affect the results is also improbable since the dispersing agent was thoroughly dried at 105°C. For future work it is recommended that a supercentrifuge (e.g. one of the Sharples type) be used to remove the barium sulphate instead of the tube type which was used in this case.

The zeta potential curves (Figure 3) are typical for inert surfaces in simple electrolytes which do not reverse the sign of charge. The zeta

potential in the salt free dispersing agent is higher than the potential in the agent plus sodium sulphate and much higher than the potential in pure sodium sulphate. Since there was no dispersing action in the pure sodium sulphate and since the maximum dispersing power in the salt free dispersing agent was greater than the maximum dispersing power in the agent plus sodium sulphate, it would seem that there might be a relation between dispersing power and zeta potential. If there is such a relation it probably is extremely complex because there is no simple relation between the curves of dispersing power and of zeta potential.

Since the zeta potential curves were typical for potentials at inert surfaces in simple electrolytes, it was assumed in the calculation of the surface charge density from equation (2) that the dispersing agent behaved as a simple electrolyte of the univalent type $\text{Na}_2\text{R} \rightleftharpoons 2\text{Na}^+ + \text{R}^-$ where R is the organic radical dinaphthylmethanedisulphonate. The points obtained from these calculations, and which are plotted in Fig. 4, very nearly follow the curves calculated from equation (3). Since equation (3) describes the course of the surface density of charge on inert surfaces in simple electrolytes, it would seem that the dispersing agent behaves as a

simple electrolyte and is ion-adsorbed on the surface of the suspended particles.

The curves of dispersing power (see Figure 1) are also suggestive of the action of simple electrolytes on hydrophobic suspensions. It will be noticed that as the concentration of the agent increases, the dispersing power increases until a maximum is reached, after which the dispersing action decreases. This parallels the fact that a hydrophobic colloid which has been flocculated by the complete removal of adsorbed electrolytes will be reprecipitated by the addition of an electrolyte, and that when the concentration of that electrolyte reaches a certain value it will be flocculated.

Evidence that the dispersing action is related to the surface density of charge is given by the comparison of the dispersing power with the surface density of charge (Figure 6). It will be seen that for the salt free agent the dispersing power is very nearly directly proportional to the surface density of charge up to the point of maximum dispersing power, after which this relation does not hold, since flocculation has begun. A similar linear relation is found for the agent plus sodium sulphate in very dilute solutions, but as the concentration increases, the curve falls off somewhat,

then rises, and falls again. This peculiar dip in the curve may be due to ion antagonism between the organic radical of the dispersing agent and the sulphate radical.

A consideration of the method of measuring the dispersing power leads to implications of a more general nature. It will be realized that the quantity termed relative dispersing power is also a measure of the stability of the dispersion. It is possible then, that the stability of a suspensoid is related directly to the surface density of charge.

Figure 2 shows that the pH of solutions of the dispersing agent varies somewhat with concentration of the agent in them. This may be due to hydrolysis. The fact that the pH of the dispersions made with these solutions does not differ much from that of the solutions themselves indicates that hydrolytic adsorption is negligible.

The surface tension of solutions of the dispersing agent is considerably reduced a short time after pouring, and is increased as time increases (see Figure 5). It may be that the initial reduction in surface tension aids in the wetting of the to-be-dispersed material by being adsorbed mechanically at the solid - solution interface and

that as time goes on, a mechanical desorption takes place which is followed by the ionadsorption discussed above. In connection with the surface tension of the solutions, it is rather interesting that the time-equilibrium values approached those taken to a constant value. Perhaps the slight stirring of the liquid-air interface effected by the movement of the ring of the tensiometer through it at short intervals of time caused the equilibrium concentration of the dispersing agent in the interface to be reached sooner than when the concentration of the dispersing agent in or out of the interface was allowed to proceed by itself.

The viscosity - concentration curve (Figure 7) of the dispersing agent is a straight line over the concentration range which was employed. This would indicate that the dispersing agent is not a highly dispersed, solvated colloid, like gelatin for example.

VI. CONCLUSIONS

A consideration of the results indicates that the following conclusions are probable.

1. The dispersing agent behaves as though it were ionadsorbed on the dispersed particles which increases the zeta potential and surface density of charge considerably above the values obtained in an ordinary electrolyte like sodium sulphate.

2. The most important factor governing the dispersing action is the surface charge density to which the dispersing power is directly proportional in dilute solutions of the agent.

VII. RECOMMENDATIONS

1. That a study be made of the stability of dispersions in ordinary electrolytes and other dispersing agents and that the relation of it to the surface density of charge be determined.

2. That the actual adsorption of dispersing agents on carbon particles be measured with the object of checking the results of the electrokinetic method. This might be done by an ultra-filtration, colorimetric method.

VIII. APPENDIX

1. CELL DIMENSIONS:

A. Depth of the cell

	Lower surface	No. of Revs.	Upper Surface	Total no. Scale divs.	Average
Left end	6.0	6	31.5	325.5	
	14.8	6	34.9	320.1	
	12.5	6	34.5	322.0	
	12.2	6	34.7	322.5	322.5
Center	2.2	6	26.2	324.0	
	2.4	6	26.5	324.1	
	3.8	6	27.3	323.5	
	3.1	6	26.7	323.6	323.8
Right end	32.4	6	6.2	324.2	
	33.9	6	7.3	323.4	
	32.2	6	5.2	323.0	
	31.6	6	5.2	323.6	323.5
				3 969.8	
					323.2

Depth = $323.2 \times 0.002 = 0.6460\text{mm}$.

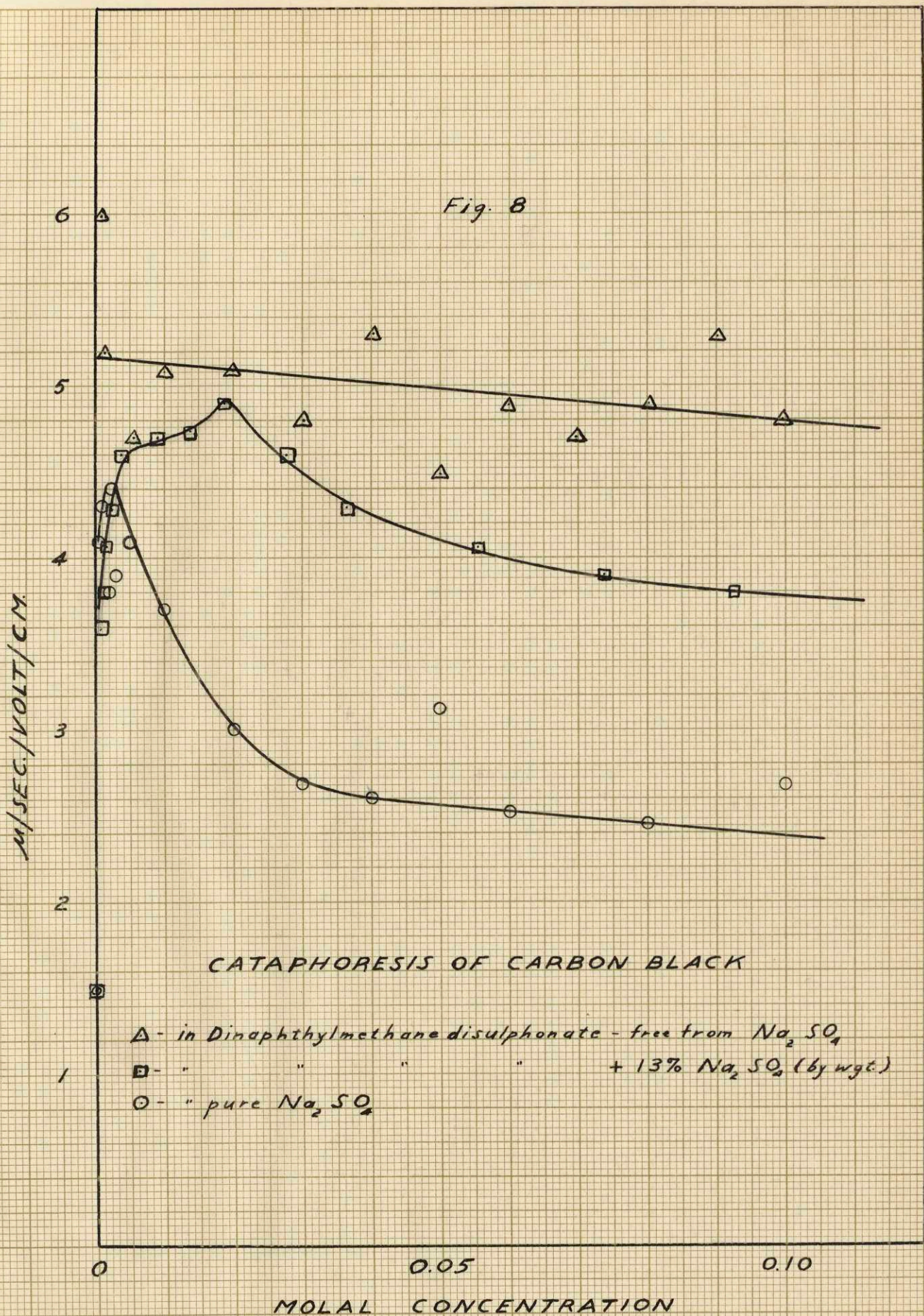
B. Width of cell = 12 mm.

C. Distance between platinum electrodes = 35mm.

D. 50 ocular micrometer divisions = 0.09mm. = 90 microns;

using 40x water immersion objective and binocular eyepiece.

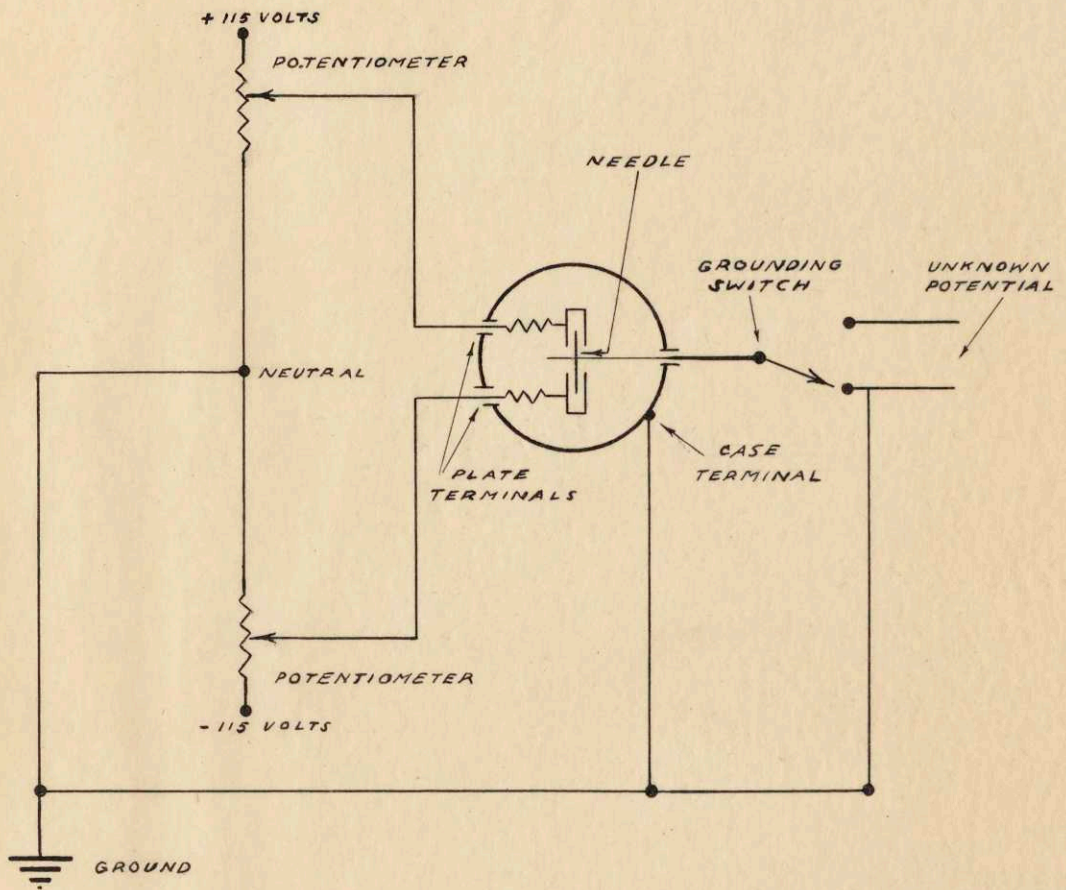
Fig. 8



CATAPHORESIS OF CARBON BLACK

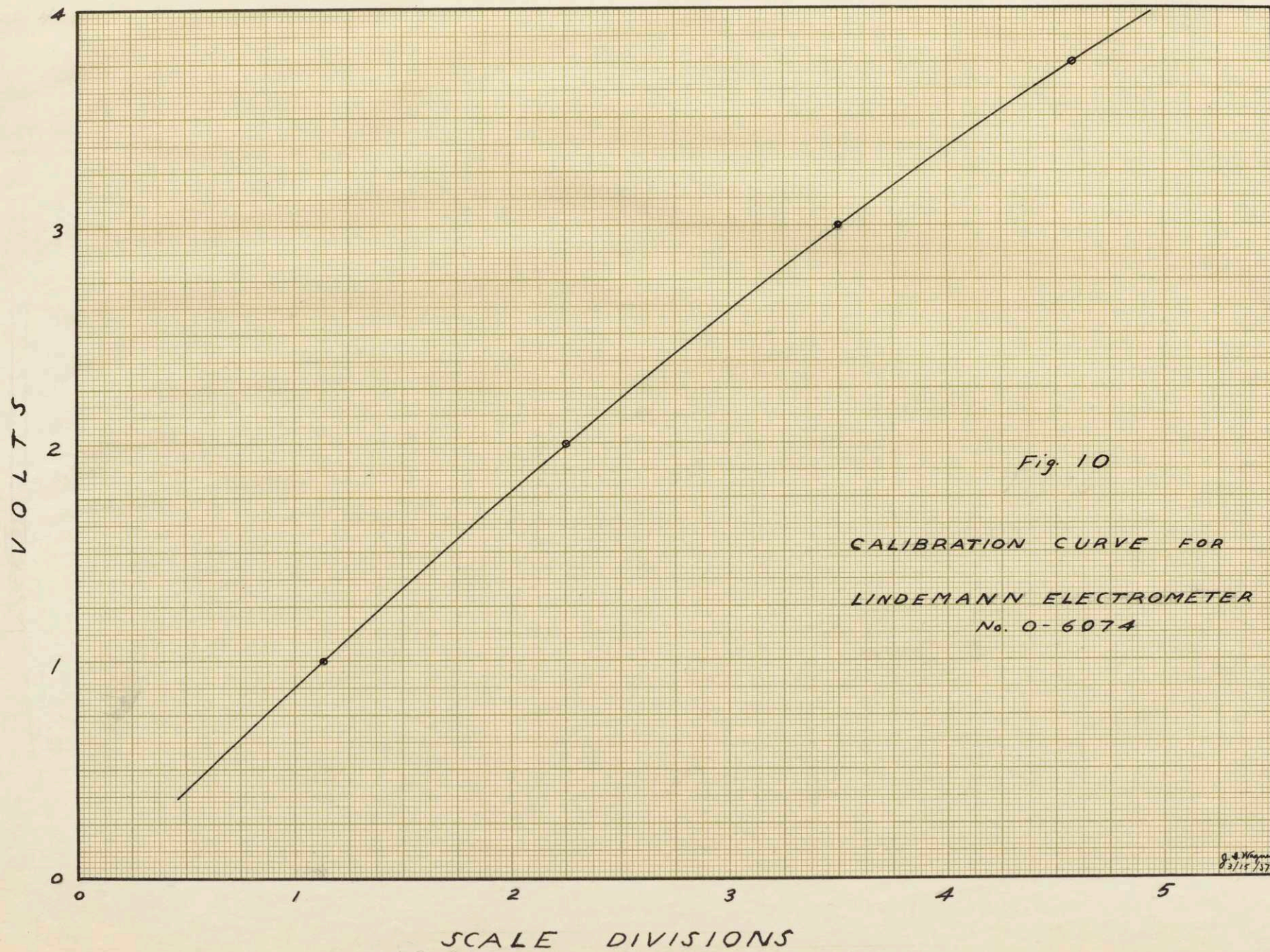
- Δ - in Dinaphthylmethane disulphonate - free from Na₂SO₄
- - " " " " + 13% Na₂SO₄ (by wgt.)
- - " pure Na₂SO₄

Fig. 9



CIRCUIT DIAGRAM - LINDEMANN ELECTROMETER

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2. CALIBRATION OF CATAPHORESIS CELL:

(1). V _{zn} in volts	(2). V _{pt} with V _{zn} = 0	(3). V _{pt} with V _{zn} = V _{zn}	(4). V _{pt} in scale divs. =	(5). V _{pt} in volts	V _{pt} = (5) <u>V_{zn}</u> (1)
			(3) - (2)		
9.90	-0.50	1.65	2.15	1.90	0.192
9.95	-0.40	1.75	2.20	1.95	0.196
9.96	-0.60	1.85	2.45	2.16	0.217
10.00	-0.70	1.85	2.55	2.24	0.224
10.00	-0.70	1.85	2.55	2.24	0.224
17.00	-0.95	3.80	4.75	3.86	0.227
17.00	-0.80	3.90	4.70	3.83	0.225
17.08	-0.75	3.90	4.65	3.80	0.223
16.95	-1.10	3.10	4.20	3.48	0.205
17.00	-0.55	4.25	4.80	3.89	0.229

$10 \overline{) 2.162}$
 0.216 av.
 or 0.22

The voltage drop /cm.in the cell =

$$V_{zn} \times \frac{0.22}{3.5} = V_{zn} \times 0.063$$

V_{zn} = Potential drop across zinc electrodes.

V_{pt} = " " " platinum electrodes.

3. CALCULATIONS =

A. Depth of stationary layer

Equation of Komagata (1)

$$y = \pm b \sqrt{\frac{1}{3} \left(1 + \frac{384}{\pi^5 k} \right)}$$

where y = depth of stationary layer from the central axis.

$$k = \text{ratio of cell width to cell thickness} = \frac{2a}{2b} = \frac{12}{0.646} = 18.6$$

$$\therefore y = \frac{0.646}{2} \sqrt{\frac{1}{3} \left(1 + \frac{1.246}{18.6} \right)} = 0.191 \text{ mm. from central axis.}$$

∴ Distance of the Stationary layer from the bottom or top of the cell = $\frac{0.323 - 0.191}{0.646}$ X depth of cell

$$= 0.204 \times 323.2 = 66.0 \text{ scale divisions on the micrometer screw.}$$

23.2

B. Cataphoretic velocity (v)

$$v = \frac{\text{observed velocity in microns/second}}{\text{voltage drop/cm. in the cell}}$$

$$= \frac{\text{observed velocity}}{V_{zn} \times 0.063}$$

C. Viscosity

$$\frac{N}{N_0} = \frac{Rt}{R_0 t_0}$$

where N = coefficient of viscosity
R = density
t = time

$$\text{At } 25^\circ\text{C, } N_0 = 0.00894$$

$$R_0 = 1.000$$

$$\therefore N = 0.00894 \times R \times \frac{t}{t_0}$$

D. Zeta potential

$$\zeta = \frac{4\pi\eta v}{D} \quad (\text{in e.s.u.})$$

1 cgs e.s.u. of potential = 300 volts

$$\therefore \zeta = \frac{4\pi \times 1000}{81.1 \times 10000} \times (300)^2 \times \eta v$$

$$= 1394 \eta v \quad \text{millivolts when } (v) \text{ is in microns/second/volt/cm.}$$

E. Surface density of charge (σ) :

$$\sigma = \sqrt{\frac{NDkT}{2\pi \cdot 1000} \cdot c_{1(2)} z_{1(2)} \left[\frac{1}{z_2} \left(e^{z_2 \frac{E\mathcal{J}}{kT}} - 1 \right) + \frac{1}{z_1} \left(e^{-z_1 \frac{E\mathcal{J}}{kT}} - 1 \right) \right]}$$

In the case of a salt of the type Na_2R , $z_2 \neq 2z_1$ and

$$\sigma = \sqrt{\frac{NDkT}{2\pi \cdot 1000} \cdot c_2 \cdot 2z_1 \left[\frac{1}{2z_1} \left(e^{2z_1 \frac{E\mathcal{J}}{kT}} - 1 \right) + \frac{1}{z_1} \left(e^{-z_1 \frac{E\mathcal{J}}{kT}} - 1 \right) \right]}$$

$$= \sqrt{\frac{NDkT}{2\pi \cdot 1000} \cdot c_2 \left[e^{2z_1 \frac{E\mathcal{J}}{kT}} + 2e^{-z_1 \frac{E\mathcal{J}}{kT}} - 3 \right]}$$

where $z_1 = 1$, with \mathcal{J} in volts, and at 25°C .

$$\sigma = \sqrt{\frac{6.06 \times 10^{23} \times 8.11 \times 1.37 \times 10^{-16} \times 298}{2 \times 3.142 \times 1000}} \cdot \sqrt{c_2} \cdot \sqrt{e^{2 \times 1 \frac{4.77 \times 10^{-10} \times \mathcal{J}}{1.37 \times 10^{-16} \times 298 \times 300}} + 2e^{-1 \times \frac{4.77 \times 10^{-10} \times \mathcal{J}}{1.37 \times 10^{-16} \times 298 \times 300}} - 3}$$

$$= 17900 \sqrt{c_2} \sqrt{e^{77.6\mathcal{J}} + 2e^{-38.8\mathcal{J}} - 3}$$

F. Adsorption curve for surface density of charge

$$\sigma = \sigma_M \beta' \frac{c}{1 + \beta'c}$$

For example in pure sodium sulphate,

at $c = 0.0005$, $\sigma = 2960$, and

at $c = 0.002$, $\sigma = 4860$; and therefore

the slope ($\sigma_M \beta'$) of the curve equals

$$\frac{4860 - 2960}{0.002 - 0.0005} = 12.66 \times 10^5$$

Taking another point; at $c = 0.03$, $\sigma = 11700$,

$$\therefore 11700 = 12.66 \times 10^5 \times \frac{0.03}{1 + \beta' \cdot 0.03}$$

$$\text{and } \beta' = 74.6$$

$$\therefore \sigma = 12.66 \times 10^5 \times \frac{c}{1 + 74.6 c}$$

DATA

1. Viscosity of the dispersing agent plus 13%

Na₂SO₄ at 25° ± 0.1°C

(1) Molal Conc.	(2) Time (t) secs.	(3) $\frac{t}{t_0} =$ $\frac{(2)}{95.5}$	(4) R/R ₀	(5) N= 0.00894x(3)x(4)
0.0000	95.5	1.000	1.000	0.00894
0.0004	95.8	1.002	1.000	0.00896
0.0009	96.3	1.005	1.000	0.00899
0.0014	96.6	1.008	1.000	0.00900
0.0018	96.9	1.010	1.000	0.00903
0.0037	96.2	1.003	1.000	0.00897
0.0092	96.2	1.003	1.000	0.00897
0.0138	97.7	1.019	1.001	0.00912
0.0184	97.8	1.020	1.004	0.00915
0.0276	99.4	1.038	1.007	0.00934
0.0368	100.2	1.046	1.009	0.00944
0.0552	102.5	1.070	1.014	0.00970
0.0736	104.5	1.090	1.018	0.00992
0.0922	106.5	1.111	1.031	0.01024

Viscosity of pure Na₂SO₄ (from International
Critical Tables) at 25°C.

0.100	0.00930
0.250	0.01038
0.500	0.01097
0.750	0.01229

2. Dispersing power:

Salt free agent			Agent + 13% Na ₂ SO ₄		
Molal conc.	Reading	% Absorption	Molal conc.	Reading	% absorption
0.0000	1.7	2.1	0.0000	1.7	2.1
0.0005	10.0	12.5	0.0004	1.7	2.1
0.001	5.0	6.3	0.0009	3.2	4.0
0.005	9.5	11.9	0.0014	3.8	4.8
0.010	10.5	13.1	0.0018	4.9	6.1
0.020	12.0	15.0	0.0037	8.3	10.4
0.030	12.0	15.0	0.0092	13.8	17.3
0.040	15.0	18.8	0.0138	14.0	17.5
0.050	15.6	19.5	0.0184	15.0	18.8
0.060	20.0	25.0	0.0276	16.3	20.4
0.070	21.7	27.2	0.0368	19.5	24.4
0.080	24.5	30.6	0.0552	19.0	23.8
0.090	15.0	18.8	0.0736	13.0	16.3
0.100	18.5	23.1	0.0922	3.5	4.4

3. Cataphoresis and zeta potential in:

A. Salt free dispersing agent

Molal conc.	V _{zn} volts	Time(secs)for particle to go 45 microns	Cataphoretic velocity(v) in μ/sec./volt/cm.	Nx10 ³	(v)xNx10 ³	ζ in milli- volts
0.0005	29.2	4.1	6.0	8.95	53.7	75
0.001	29.2	4.7	5.2	8.96	46.6	65
0.005	29.5	5.2	4.7	9.01	42.3	59
0.010	29.5	4.7	5.1	9.08	46.3	64.5
0.020	29.7	4.7	5.1	9.22	47.1	66
0.030	29.8	5.0	4.8	9.35	44.8	62.5
0.040	29.7	4.5	5.3	9.49	50.3	70
0.050	29.7	5.4	4.5	9.63	43.3	60
0.060	29.7	4.9	4.9	9.77	47.8	67
0.070	29.7	5.1	4.7	9.91	46.5	65
0.080	29.7	4.9	4.9	10.05	49.3	69
0.090	29.8	4.5	5.3	10.18	54.0	75
0.100	29.8	5.0	4.8	10.32	49.5	69

B. Dispersing agent + 13% Na₂SO₄

Molal conc.	V _{zn} volts	Time(secs) for particle to go 45 microns	Cataphoretic velocity(v) in μ /sec./volt/cm.	Nx10 ³	(v)xNx10 ³	ζ in milli-volts
0.0004	28.3	7.1	3.6	8.94	32.2	45
0.0009	29.3	7.4	3.3)	8.95	36.7	51
	29.4	5.0	4.9) 4.1			
0.0014	29.8	8.0	3.0)	8.96	34.1	47.5
	29.6	5.4	4.5) 3.8			
0.0018	28.8	5.8	4.3	8.97	38.6	54
0.0037	29.9	5.3	4.6	8.99	41.3	58
0.0092	28.8	5.1	4.9)	9.06	43.0	60
	28.4	5.5	4.6) 4.75			
0.0138	28.6	5.2	4.8)			
	28.4	5.5	4.6) 4.7	9.13	42.9	60
0.0184	29.4	5.0	4.9	9.19	45.1	63
0.0276	29.2	5.6	4.4)			
	28.6	5.1	4.9) 4.6	9.32	42.8	60
0.0368	29.1	5.7	4.1)			
	29.1	5.4	4.5) 4.3	9.44	40.6	57
0.0552	29.4	6.0	4.0)			
	29.8	5.8	4.1) 4.05	9.70	39.3	55
0.0736	29.4	6.4	3.8)			
	29.5	5.8	4.1) 3.9	9.95	38.8	54
0.0922	29.4	6.8	3.6)			
	29.3	6.1	4.0) 3.8	10.21	38.8	54

C. Pure sodium sulphate

Molal conc.	V _{zn} volts	Time(secs) for particle to go 45 microns	Cataphoretic velocity(v) in μ /sec./volt/cm.	Nx10 ³	(v)xNx10 ³	ξ in milli-volts
0.0005	29.8	5.6	4.3)	4.1	8.94	36.7
	29.3	6.3	3.9)			
0.001	29.8	5.6	4.3	4.1	8.94	38.4
0.002	29.8	6.4	3.8			
0.0025	29.8	5.5	4.4	4.1	8.95	39.4
0.003	29.8	6.1	3.9			
0.005	29.8	5.6	4.3)	4.1	8.96	36.7
	29.3	6.3	3.9)			
0.010	29.8	6.2	3.8)	3.7	8.975	33.2
	29.7	6.7	3.6)			
0.020	28.8	8.2	3.0	3.1	9.01	27.2
0.030	29.2	8.7	2.8			
0.040	29.2	9.6	2.6	3.1	9.08	23.6
0.050	29.8	6.0	3.9)			
		29.2	10.7	2.3)	3.1	9.12
0.060	29.2	10.6	2.3)			
		50.0	5.2	2.7)	2.5	9.16
0.080	29.4	10.2	2.4)			
		50.0	5.7	2.5)	2.45	9.23
0.10	29.5	8.5	2.8)			
		50.0	5.5	2.6)	2.7	9.30

4. Surface density of charge calculated from equation (3)

A. In the salt free agent.

Molal conc. of Disp. agent	Molal conc. of Na_2SO_4	Surface density of charge e.s.u./cm. ²
0.001	-	1840
0.010	-	16500
0.030	-	40200
0.050	-	56400
0.100	-	80900

B. In the dispersing agent + sodium sulphate

0.0009	0.0014	2300
0.0092	0.139	17200
0.0276	0.0417	33600
0.0552	0.0833	43900
0.7360	0.1111	47700

C. In pure sodium sulphate

-	0.001	1178
-	0.005	4500
-	0.010	7250
-	0.030	11700
-	0.050	13400
-	0.080	14580

5. Surface density of charge calculated from equation (2).

A. Salt free dispersing agent

Molal conc. of disp. agent	Molal conc. of sodium sulphate	Total conc. of electrolyte	Zeta pot. in Milli-volts	$\frac{(2) \times 77.6}{1000}$	$e^{(3)}$	$ze^{-38.8 \times (2)}$	$(4) + (5) - 3 \sqrt{(6)}$	$\sqrt{(1)}$	$\sqrt{(1)}$	$\sigma = (7) \times (8) \times 17900$
		(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)	
0.0005	0.0000	0.0005	75	5.82	370	negligible	367	19.1	0.0224	7640
0.001	0.0000	0.001	65	5.05	156	"	153	12.4	0.0316	7020
0.005	0.0000	0.005	59	4.58	98	0.2	95.2	9.8	0.0706	12400
0.010	0.0000	0.010	64.5	5.00	150	negligible	147	12.1	0.100	21700
0.020	0.0000	0.020	66	5.12	167	"	164	12.8	0.141	32300
0.030	0.0000	0.030	62.5	4.85	128	"	125	11.2	0.173	34400
0.040	0.0000	0.040	70	5.44	230	"	227	15.1	0.200	54000
0.050	0.0000	0.050	60	4.65	105	"	102	10.1	0.223	40300
0.060	0.0000	0.060	67	5.20	180	"	177	13.3	0.245	58800
0.070	0.0000	0.070	65	5.05	156	"	153	12.4	0.264	58600
0.080	0.0000	0.080	69	5.35	210	"	207	14.4	0.282	73000
0.090	0.0000	0.090	75	5.82	370	"	367	19.1	0.300	102000
0.100	0.0000	0.100	69	5.35	210	"	207	14.4	0.316	81500

B. Dispersing agent plus sodium sulphate

Molal conc. of disp. agent	Molal conc. of sodium sulphate	Total conc. of electrolyte	Zeta Pot. in Milli-volts	(2) $\frac{x \ 77.6}{1000}$	(3) $e^{(3)} \ 2e^{-38.8x(2)}$	(4) + (5) - 3	$\sqrt{(6)}$	$\sqrt{(1)}$	$\sigma = (7)x(8)x$	17900
		(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)	
0.0004	0.0002	0.0006	45	3.48	32.5	0.3	29.8	5.45	0.0245	2390
0.0009	0.0005	0.0014	51	3.97	53.0	0.3	50.3	7.1	0.0374	4750
0.0014	0.0007	0.0021	47.5	3.69	40.0	0.3	37.3	6.1	0.0468	5120
0.0018	0.0009	0.0027	54	4.17	65.0	0.25	62.25	7.9	0.0520	7360
0.0037	0.0019	0.0056	58	4.47	87.6	0.2	84.8	9.2	0.0748	12300
0.0092	0.0047	0.0139	60	4.65	105.	0.2	102.2	10.2	0.118	21500
0.0138	0.0071	0.0208	60	4.64	104	0.2	101.2	10.05	0.145	26100
0.0184	0.0094	0.0278	63	4.88	132	0.2	129.2	11.4	0.167	34100
0.0276	0.0141	0.0417	60	4.63	103	0.2	100.2	10.0	0.204	36500
0.0368	0.0187	0.0555	57	4.62	102	0.2	99.2	9.95	0.236	42000
0.0552	0.0281	0.0833	55	4.25	70.1	0.25	67.35	8.21	0.288	42400
0.0736	0.0375	0.1111	54	4.20	67.0	0.25	64.25	8.02	0.333	47700

C. In pure sodium sulphate.

Molal conc. of disp. agent	Molal conc. of sodium sulphate	Total conc. of elect- rolyte	Zeta pot. in Milli- volts	$\frac{(2) \times 77.6}{1000}$	(3) e	$-38.8 \times (2)$ 2e	$(4) + (5) - 3$	$\sqrt{(6)}$	$\sqrt{(1)}$	$\sigma =$ $(7) \times (8) \times$ 17900
		(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)	
0.0000	0.0005	0.0005	51	4.05	57.6	0.3	5.49	7.40	0.0224	2960
0.0000	0.001	0.001	53.5	4.15	63.8	0.25	61.05	7.83	0.0316	4430
0.0000	0.002	0.002	47	3.68	39.6	0.3	36.9	6.06	0.0447	4860
0.0000	0.0025	0.0025	55	4.26	70.9	0.2	68.1	8.25	0.050	7380
0.0000	0.003	0.003	49	3.78	44.0	0.3	41.3	6.43	0.0548	6320
0.0000	0.005	0.005	51	3.97	53.0	0.3	50.3	7.10	0.0706	8970
0.0000	0.010	0.010	46	3.59	36.2	0.3	33.8	5.81	0.100	10400
0.0000	0.020	0.020	38	2.94	18.9	0.5	16.4	4.05	0.141	10200
0.0000	0.030	0.030	35	2.82	16.8	0.5	14.3	3.78	0.173	11700
0.0000	0.040	0.040	33	2.55	12.8	0.6	10.4	3.22	0.200	12900
0.0000	0.060	0.060	32	2.47	11.8	0.6	9.4	3.06	0.245	13400
0.0000	0.080	0.080	31.5	2.45	11.6	0.6	9.2	3.03	0.282	15300

6. pH of solutions and dispersions.

A. Salt free dispersing agent

Molal conc.	pH of solution	pH of dispersion
0.0005	6.61	6.26
0.001	6.46	5.62
0.005	4.63	4.66
0.010	4.44	4.75
0.020	4.82	4.98
0.030	4.51	5.01
0.040	4.92	4.96
0.050	5.32	5.10
0.060	6.03	5.21
0.070	6.11	5.70
0.080	6.12	5.70
0.090	6.34	5.84

B. Dispersing agent + sodium sulphate

0.000	5.93	6.51
0.0004	7.39	7.41
0.0009	7.30	7.41
0.0018	7.30	7.94
0.0037	7.60	7.50
0.0092	7.93	7.70
0.0138	7.78	7.72
0.0184	9.00	8.35
0.0276	8.88	8.63
0.0368	8.98	8.88
0.0552	9.00	8.85
0.0736	8.54	8.46

7. Surface tension of salt free dispersing agent
in dynes/cm. Equilibrium surface

Molal conc.	Reading taken to constant value	Equilibrium surface			
		2 hrs.	3 hrs.	14 hrs.	12 hrs.
0.000	74.4	74.4			
0.0005	74.7	61.2	72.2	74.9	75.4
0.001	74.4	61.2	75.6	75.4	75.1
0.005	73.0	70.1	69.5	73.	74.8
0.010	73.1	73.5	74.4	73.9	73.7
0.020	72.8	58.5	61.3	68.1	72.4
0.030	73.0	63.5	69.0	68.4	69.4
0.040	73.2	58.5	61.9	67.4	71.3
0.050	73.0	61.9	63.7	63.6	68.3
0.060	73.0	62.3	67.6	70.2	71.6
0.070	73.1	55.1	57.2	61.3	68.9
0.080	72.8	70.0	72.0	72.3	72.2
0.090	72.8	60.8	64.3	67.5	69.5
0.100	72.0	59.5	66.7	69.3	69.5

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