# ON THE INTERCHANGE OF ELECTRICITY BETWEEN SOLIDS, LIQUIDS AND GASES IN MECHANICAL. **ACTIONS\***

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**ABSTRACT.** When liquids or solid particles are subjected to various mechanical processes. such as frictional force, spraying, splashing, raining or blowing, striking against each other, etc , electric charges are developed in the substance, in the apparatus used in the mechanical process in and the surrounding medium. Similarly, some gas jets produce positive charge and others negative charge on the nozzle or on the plate on which they impinge. We have no complete picture for any one case to show how the interchange of electricity takes place between the apparatus, substance and the medium; consequently, experiments were undertaken with a number of substances, initially uncharged and initially charged, to supplement the observations made by previous workers. These show, as one should expect, that -

(1) The total charge in the substance, in the apparatus (or escaping into earth through the apparatus) and the medium in any mechanical process is zero. While the total remains zero, the distribution between the apparatus, substance and the medium (positive and negative ions) shows wide variations according to the process employed and the substance used. If the operation produces in any case equal number of positive and negative ions in the medium, the substance and the apparatus either collectively or separately remain uncharged.

(2) The charge developed depends on the violence of the mechanical action. Only a part of the work done in such processes is converted into electricity.

If the existence of an electrical double-laver between the substance and the medium is assumed, the jous in the medium come from both layers of the double-layer, but generally more from the layer situated on the molecules of the medium. The more violent is the process the greater is the number of ions dislodged from the layer situated on the molecules of the substance.

If a substance has an initial charge, this charge does not play any part in the interchange of electricity between it and the medium. The initial charge adheres to the substance during the process of disruption.

The exchange of electricity between substance and medium in all mechanical actions can be explained on the assumption of an electrical double-laver.

When increasing quantities of NaCl or HCl are dissolved in distilled water, the charge in the substance changes very sharply from positive to negative at a very low concentration The distribution in such cases of charge in the substance, in the apparatus, and in the air in the shape of positive and negative ions is discussed particularly at the stage when the transition from positive to negative takes place. When the substance passes through the transition point, a reversal of the double-layer takes place

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The interchange of electricity brought into play in some of the mechanical actions in the at nosphere is briedy considered and it is indicated that some of the earlier calculations need revision.

The various factors which contribute to the generation of charge in such processes and the possible genesis of the electrical double-layer on the electronic theory of matter are briefly discussed,

#### PART I-EXPERIMENTAL, DATA

#### I. introduction

A number of workers have carried out experiments on the electric charge developed when liquid drops (organic or inorganic) and solid particles are subjected to mechanical processes of various types. These involved spraying or splashing of liquids and raining or blowing of dust particles and in most eases attempt was made to explain the observed results on the assumption of an electrical double-layer. Unfortunately, we have no complete information for any case ; those udio have measured the charge on the substances have in most cases not recorded the charge in the medium or in the inslrumcnts employed and those who have recorded the charge in llie medium have not noted the charge in the substances. Consequently, the precise rôle of the double-layer in the interchange of electric charge between the substance and medium when the former is subjected to breaking or friction has received no satisfactory elucidation.

I'he colloidal chemists assume electric charge to be a fundamental propeily of the colloidal particles. The existence of an electrical double-layer is assumed and the charge is supposed to have arisen as a result of 'slipping' of this doublelayer. According to Mukherjee,  $<sup>1</sup>$  the inner layer forms a firmly adhering charge.</sup>

A simple experiment shows that aerosols have not necessarily a charge initially. If a jet of condensed steam is allowed to escape between two metal plates, placed 20 cm. apart, and charged to a potential difference of 50,000 volts, the steam is found to remain unaffected. If, 011 the other hand, the jet is subjected to a beam of X-rays, before it escapes between the two plates, the condensed steam is found to be highly charged and is attracted to one or the other of the plates according to its polarity. If the vapour particles in the jet be regarded as aerosols, then clearly the experiment shows that initially they are not charged. In other words, no slipping of the double-layer has occurred, and each particle is, therefore, uncharged.

Two fundamental questions arisc—

*[a)* Is the assumption of the existence of a double-layer essential for the explanation of the development of ekctinc charge in all mechanical processes *?*

(b) What is the precise rôle of the double-layer in the development of such charges ?

We propose to examine both these questions with the aid of data obtained by previous workers on the subject and those obtained by the author with the assistance of a co-worker, Mr. R. D. Godbole, in order to supply the missing link.

#### 2. A V A I L A B L E D A T A

In 1892 Lenard<sup>2</sup> showed that water jets or drops striking against an obstacle arc charged positively, while the surrounding air becomes negatively charged ; the effect being reduced considerably by the presence of even slight impurities in the liquid used. A very important contribution was made by J. J. Thomson<sup>3</sup> in 1894 when he brought to notice that the electrification of drops of distilled water striking against a metal surface suffers a reversal when hydrogen is substituted for air. In the literature of colloidal particles we find that substances of higher dielectric constant with respect to the medium get positively charged. This is clearly not true for breaking water drops, for, water has a higher dielectric constant both with respect to air and hydrogen, and yet in air the drops get positively charged and in liydiogeii negatively charged.

TIiomson showed that solutions such as phenol, pyrogallic acid, fluorcscene, whose drops are electrified in the same way as those of water are affected by hydrogen in the same way as drops of water are affected, *i.e.*, drops which would be positively charged in air would become negatively charged in hydrogen. On the other hand, the effect of substituting hydrogen for air in the case of substances like methyl violet, rosaniline, potassium permanganate, whose drops show negative electrification in air is to increase this negative electrification. If experiments are made with different gases the magnitude of the charge is found to depend largely on the amount of gas absorbed by the drops, by which they are surrounded. Thus, water falling through sulphuretted hydrogen gives very little charge, while water falling through its own vapour gives no effect. The temperature of the substance is also a factor of considerable influence. Thus, while water at  $15^{\circ}$ C, allowed to strike a plate gave a deflection of 230 scale divisions in the electrometer, that at 75<sup>°</sup>C and 95<sup>°</sup>C produced deflections of 390 and 420 divisions respectively.

Evc<sup> $\dagger$ </sup> has made available to us a considerable amount of data in regard to the ionisation produced in the air by spraying "different liquids. Unfortunately, he kept no record of the charge in the substance or in the apparatus. His results show that generally both positive and negative ions are produced by spraying, and that the number of the ions and the ratio of the negative to positive ions are characteristic of the substance sprayed. He also found that surface tension and the extent to which liquids are volatile do not appear to influence the relative ionisabilities of different substances. Busse<sup>5</sup> showed that the production of positive ions takes place in a greater quantity in the case of spraying of water (the

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ratio of positive ions to negative ions being about *2 :* 3) than in the case of strik ing and bubbling processes (the ratio in these cases being roughly  $i : 15$ ).

Nolan $<sup>6</sup>$  investigated the dependance of the separation of electrical charge on</sup> the surface produced, by measuring the charge on various sizes of drops produced by splashing and spraying water. If the charge developed  $(Q)$  be expressed in the form  $Q = \sigma A$ , where A is the area of the surface, then  $\sigma = 2.5 \times 10^{-3}$  e.s.  $u./cm.^2$ 

Rudge<sup>7</sup> showed that when dust particles are blown, the sign of the charge depended on the nature of the substance,  $c.g.,$  acidic bodies and nonmetallic elements gave positively charged clouds, while basic bodies and metallic elements gave negatively charged clouds. The electrification of dust and air was in general, but not in every case, found to be of opposite sign. Deodar's tests<sup>\*</sup> on graded dust of different sizes showed that, other things being equal, the charge developed by blowing increased very rapidly as the size decreased. The charge also depended on the nature of the substance used. I'or instance, while NaCl and KCl were equally efficacious, NaNO<sub>3</sub> was about  $4\frac{1}{2}$  times more efficacious in producing charge than  $KNO<sub>a</sub>$ .

#### **r U R PT K K** *n* **X K R <sup>1</sup>** *M* **K N** *r* **S**

It will be seen from the above review that although a considerable series of experiments have been carried out on the subject, no author has recorded the charge in the substance, in the air and in the apparatus used for producing mechanical action. For instance, when water is sprayed, electric charge is developed in the sprayer, in the sprayed water drops, as well as in the surrounding air. Except in the case of water, we have no quantitative values of tlie charge in the substance and that in the medium in any particular operation. For water drops broken in a vertical blast of air, Simpson<sup>®</sup> recorded the charge both in the substance as well as in the air. In order to get connected records of the total charge developed in such processes, it became necessary to carry out further experiments in which arrangements were made for recording the charge in the substance, in the air, as well as in the apparatus.

The mechanical processes employed for the development of charge were the following :—

In the case of liquids— \*

- (a) Drops broken in a vertical blast of air,
- *(b)* Drops striking a plate,
- $\epsilon$ ) Jet striking a plate,
- (d) Two jets striking against each other in the air,
- (e) Spraying by an atomiser.

In the case of solid particles-

- $(a)$  Particles raining through sieve and received on a plate,
- $(b)$  Particles blown in the form of a cloud,
- $(c)$  Particles slipping down two inclined planes and striking against each other in the air.

In the case of gas-

- (a) A gas jet striking a plate,
- $(b)$  Two gas jets striking against each other.

In most cases the mechanical process was made automatic so that the same effect could be repeated as many times as desired.

The experimental arrangement usually adopted for spraying experiments is as shown in Fig. 1. For most of the experiments the receiver used was made out of an aluminium sheet. It was of a rectangular shape (6 in.  $\times$  3} in.  $\times$  34 in.) with a circular tube of diameter  $I_1^1$  in., length  $2$  in., fixed in the middle of a vertical It had a top cover which could be removed when necessary. Suitable side modification is made in the receiver when experiments are made with jets or solid particles.



The substance subjected to the mechanical process is received in an insulated receiver which is connected to the needle of a Dolezalek electrometer, the quadrants being given equal and opposite potentials. The mechanical process occurs in an enclosed space, the air being subsequently drawn through an Ebert's counter, and the number of both positive and negative ions counted in two separate experiments.

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When it is desired to measure also the charge on the apparatus, the receiver and the electrometer are earthed, the receiver is disconnected and the apparatus is joined to the electrometer.

The observations arc taken in the order as described below.

Taking, for instance, the case of water sprayed, the spraying is done automatically by a motor blower. The nozzle of the atomizer is surrounded on all sides by the receiver, so that all the sprayed particles are collected by the receiver. A definite quantity of water, say i c.c., is sprayed every time and tests are made to see that the same effect is reproduced every time.

**(t)** First with a given **(o)** positive and (b) negative potential being applied instantaneously to the central rod of the Kbert's ion-counter, two leakage cm ves are drawn, with the fan working in the two cases.

(2) A definite quantity, say  $\tau$  c.c., of water is sprayed.

(3) The charge on the water collected in the receiver, as recorded by the Dolezalek electrometer is noted.

(4) The central rod of the ion-counter is given a definite positive potential.

(5) The air from the receiver is sucked in through the counter, by making

the fan of the counter work, and a leakage curve is drawn.

(6) The drop in potential, as given by the two curves  $(5)$  and  $(1)$ , gives, with the aid of the usual formula for Ebert's counter, the number of negative ions produced in the air by spraying i c.c. of water. Since both tlie leakage curves are equally affected by the ions normally present in air, these are automatically eliminated from the counting.

(7) With the receiver and the central rod momentarily earthed, 1 c c. of water is again sprayed, and the charge acquired by the receiver, as recorded by the electrometer, noted.

(8) The central rod of the counter is now given a definite negative potential.

(9) The air from the receiver is sucked in and a leakage curve drawn.

(10) The drop in potential as shown by (9) and (1), gives the number of positive ions in the air as a result of Spraying i c.c. of water.

(t i) The receiver is earthed and disconnected, the apparatus is joined to the electrometer and the charge developed in it is noted.

For these experiments, both the Dolezalek electrometer and Ebert's ion-counter (with its Woulf's electrometer) are calibrated, the capacities of both and of the accessories being determined by means of a standard condenser.

## PART II— S U B S T A N C K S I N I 'J'l A *h L \* UNCHARGED

4. We have to consider two distinct cases— substances may be initially charged or uncharged and it is best to treat them separately. We first take up the case of substances which arc initially uncharged.

When a substance is subjected to mechanical action, we find electric charge in the apparatus employed to produce mechanical action as well as in the substance and the medium. For instance, water passing through the nozzle of the atomizer gives negative charge to the atomizer and itself acquires a positive charge. A further development of charge takes place while the jet breaks into fine drops and falls into the receiver, owing to the dislodgment of ions which escape into the medium. Thus if x is the charge developed in the apparatus, the charge in the substance when it just comes out of the apparatus is  $-x$ . If  $\gamma$  is the resultant charge given to the mediuiri as a result of dislodgment of ions, then the charge in the substance due to this process is  $-y$ . We thus have the following distribution of charges :





FIGURE 2

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If initially the substance as well as the medium arc uncharged, we should expect the algebraic sum of the total charge developed in the apparatus, in the substance and the medium to be equal to zero in any mechanical process of this kind. In any single experiment it is, however, not easy to prove this law (paantitatively. For it is an extremely difficult matter to collect all the charges developed and measure them accurately. We can illustrate this by reference to a simple experiment. If distilled water is sprayed (by a Uc Vilbiss Atomizer No 15) the charges in the sprayer and the receiver are found to vary as shown in Fig. 2. The charge developed per c.c. of water sprayed is approximately as indicated below :

> In the water  $= +1.7$  e.s.u. In the sprayer  $= -0.5$  e.s.u. In  $air = -1$   $r_1$  e.s.u.

Within the limit of experimental error, therefore, the total charge is zero. The negative charge is slightly less than the positive charge. This may be partly due to the fact that all the ions arc not caught by the Ebert's apparatus. The actual magnitude of the charge also varies widely according to the process employed. Thus 1 c.c. of water striking a plate gets a charge of  $\sigma$ 234 e.s.u., while 1 c.c. of water broken in a vertical blast of air gets only 0.023 e.s.u. This difference is largely due to the difference in the intensity of mechanical action, and, therefore, also to the differences in the amounts of new surface developed in the two processes. A jet striking a plate breaks into a large number of small drops, while a drop broken in a vertical blast of air is broken into two or three drops only, so that in the former case the new surface developed is considerably more than that in the latter. Furthermore, the surfaces in action when drops strike against a metal plate are different from those in action when drops break in the air. In the former case besides water and air a metal plate is involved.

#### '5. R K I, A 'r T O N S H I P B E T W E E N W C) R K D O N E A N D C II A R *G 1%* PRODUCED

The charge in the substance is equal and opposite to the sum total of the charges in the apparatus and the medium.

The whole of the work done in producing mechanical action is not utilised in the generation of charge. **A** part of it is utilised in producing motion and mechanical disruption and another part in working against friction, or, in other words, in the generation of heat. If the total work done is W, and if that part of it which is utilised in producing motion, mechanical disruption, and production of heat be  $W_0$ , then  $W-W_0$  is the work done in producing electric charge. If Q is the charge developed in the substance, then clearly

$$
W-W_0=AQ^2,
$$

where A is a constant, which is dependent on the substance used and the mechanical process employed.

While it is comparatively easy to determine W, it is not always easy to determine  $W_0$ . It is not, therefore, always possible to obtain quantitative relationships and we have in many cases to remain satisfied with the qualitative law, which is well recognised, that the charge in the substance, as well as the number of ions of either sign in the medium, which are developed in the mechanical process, depend on the intensity of the mechanical operation.

How the charge developed in the substance increases with the violence of the mechanical action can be illustrated by a simple experiment. If we have a single downward jet striking an insulated plate, the work done can be varied by altering the head of water in the tube. The results obtained in an experiment of this kind are given in table I. The jet was produced by a tube having a narrow orifice of diameter o.2 mm. The pressure head in the tube was varied and the orifice was placed at a height of about 8 cm, above the bottom of the receiver.

| Pressure head<br>above the orifice<br>in cm. of water | Kinetic energy per e.c.<br>at the instant of break-<br>in e.g s units (approx.). | Charge per<br>e.c. in<br>c. s. u. |  |  |
|---|--|-----------------------------------|--|--|
| 4 <sup>2</sup>  | 49,050   | 0.15                              |  |  |
| 56.5  | 63,275   | 0.15                              |  |  |
| 59  | 05,725   | 0.18                              |  |  |
| 66  | 72,595   | 0.53                              |  |  |
| 75  | 81,425   | 0.30                              |  |  |
| 83  | 89,270   | 0.37                              |  |  |
| 92.5  | 98,590   | 0.11                              |  |  |
| 102   | 1,07,910   | 0.39                              |  |  |

TABLE I

Distilled water (single jet)

The experiment shows roughly that when the energy at the instant of break was doubled, the charge produced increased about four times. The charge produced depends also to a certain extent on the nature of the surface on which the jet falls. This will be clear from Fig.  $3$ .



Jet of distilled water falling on clear receiver and on surface covered with different substances

#### FIGURE 3

Similarly when two jets strike against each other, the chaige produced depends not only on the height of the liquid columns but also on the diameter of the jet. With two jets produced by liquid columns each 70 cm. high, the following figures were obtained :

#### TABLE II

Distilled water (two jets striking at angles between 22° to 25° and placed about 8 cm. above receiver)



With increase in the diameter of the bores the size of drops produced is increased and, consequently, there is a decrease in the charge produced.

Table III gives the charge obtained in the substance as well as in the medium when I c.c. of water is subjected to the processes indicated therein.

#### TABLE III

#### Distilled Water



\* This ratio decreases to 1:1.6, if the rate at which a given quantity of water is sprayed is increased.

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Table IV gives a comparison of charge developed in other substances.

#### TABLE IV



Charge in substance per  $c.c.$  in c  $s.u.$ 

The charge developed per c.c. is greater when a substance is sprayed than when a jet strikes the receiver ; also it is greater when drops are broken by a jet striking a receiver than by two jets striking against each other.

#### 6. 'r n n k h k c 'r r i c a *h <sup>t</sup> ) <sup>o</sup>* u t, k - h a y k r

If we assume the existence of a double-layer, the positive charges, in the case of a water drop in contact with air will, according to the usual conception, be situated upon the molecules of water, and the negative charges upon those of the medium, *i.e.*, air. Such a double-layer is illustrated in Fig. 4. When the two layers are nest to each other, the drop as a whole shows no charge. If there is a slip in the double-layer due to friction or other causes, the drop acquires a positive charge and the medium a negative charge. On the other hand, air bubbling through water gets negatively charged, for precisely the same reason, namely, the slipping of the double layer.



 $FIGURE +$ 

From the fact that when this drop is broken, we get ions of both signs in the air, we can immediately conclude that ions come out from *both* the layers. Moreover, from the fact that the negative ions arc always in excess of the postive ions, we can conclude that ions come out more easily from the outer layer than from the inner layer.

When water or any other liquid comes through a metal tube, such as the tube of the atomizer or a jet, how does the tube get charged? We must assume an electrical double-layer to exist at the interface between the liquid and the metal tube The moving liquid may produce a slip in the layer, and thus make the tube and the liquid get equal and opposite charges. There is also the "collector" action. Just as in a Kelvin water-dropper, the issuing drops in an electric field make the vessel get a charge opposite to those of drops, so also when liquid conies out through a rnetal tube, the tube must get a charge opposite to that of the drops if there is a pre-existing electric field, or the charges in the drops create such a field.

It is not easy to describe in simple language as to which of the processes mentioned in table III is more violent. When air bubbles through water, it is .more or less a case of slipping; so that we get in the air ions from the layer adjoining the air surface, that is to say, negative ions. When two jets of water strike against each other in the air, the process is slightly more violent. But in this case three factors contribute to the production of charge both in the substance and in the air. In the first place, the slipping of the double layer which occurs when water moves through the tubes makes the water jets positively charged. Secondly, when the jets break in the air into drops, they acquire further positive charge and give the excess negative charge to the air. Finally, when the drops strike against the receiver, the positive charge is further augmented, the excess of negative charge being given to the air. When drops are broken in a vertical blast of air, all the above three factors contribute to the production of charge. There is, however, the following difference ' in tlie two cases;—

(1) The slipping of the double-layer during movement through the tubes gives to the water jets much greater positive charge than to the drops which come out slowly one after another,

(2) The drops formed per c,c. are slightly more in number when two jets strike against one another than when drops are broken in a vertical blast of air. In the former case, action is between water and water and in the latter, between air current and water. The result is that in the former case, both the substance and the medium (as determined by total of positive and negative ions) receive greater charge, but in the latter the air current dislodges a greater proportion of positive ions so that the ratio of positive to negative ions is greater in the latter than in the former case.

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When a jet is directed against a receiver, the action is more violent than when two jets strike against each other with the result that the charges developed both in the substance and the medium are very much greater ; the ratio of the positive to negative ions is, however, very nearly the same in the two cases. W ith an atomizer the process is very violent and ions arc dislodged from both the layers and there is only a very slight excess of negative ions.

The fact that in these experiments, there is always an excess of negative ions suggests that there is one layer from which ions are more easily removed than from the other. This clearly must be the layer or the charges situated upon the molecules of the medium, that is to say, the air in this case.

In the case of the substances given in table V, the number of positive and negative ions in the air arc found to be very nearly equal, and the substances to develop very little charge. The data regarding charge in the air given in the table are those obtained by Eve converted into electrostatic units. These results were generally coufirmed by independent experiments.



TABLE V

(Sprayed by atomizer)

Such substances as amyl acetate, chloroform, aldehyde, acetic acid, also give equal number of positive and negative ions in the air on being sprayed, and acquire very little charge themselves.

If there are equal numbers of positive and negative ions in the air, the *substance and the apparatus either collectively or separately remain uncharged, however violent is the mechanical process to which the substance is subjected.*

If the double layer be supposed to exist in these cases it is clear that the medium must have derived ions from both the layers with equal ease. The inside layer in these cases is not more firmly adhering than the outside layer.

Substances generally get positively or negatively charged according as there is an excess of negative or positive ions in the air.

This result is true only in those cases where the charge acquired by the apparatus is small compared with the charge in the substance and in the air. This result was firmly established in the case of water in Simpson's experiments. In the qualitative experiments of Rudge, he came across some contradictory results showing that the substance and the medium had charge of the same On careful scrutiny of his experiments, it appears that when dust is sign. blown, or allowed to rain through a sieve, the blower or the sieve as the case may be, gets charged. The charges in the dust particles, in the air, and in the blower (or sieve) are equal to zero collectively. In those cases where the number of ions of both signs in the medium is very small or where the number of ions of one sign is very nearly equal to the number of ions of the other sign, often contradictory results are recorded in the sense that the substance is sometimes found to be positively charged and sometimes negatively charged. These appear to be due to errors in the experiments. Even the same chemical taken from two different bottles gives different results on account of slight differences in the impurities. Unless the samples are taken from the same source and the apparatus is carefully protected from stray electrification by earthed covers, and also sources (sources of ionisation) which might produce such electrification are carefully eliminated, these experiments often prove to be very elusive.

In the case of substances given in table VI the charge in the substance is of opposite sign to that of the resultant charge in the medium, except for turpentine and acetylene tetrachloride.

|                                      | Charge derived from 1 c.c. of the substance in e.s.u. |                |           |            |  |  |  |  |
|--------------------------------------|---|----------------|-----------|------------|--|--|--|--|
| Substance.                           | Charge in the<br>substance.                           | Charge in air. |           |            |  |  |  |  |
|                                      |   | Posi           | Negative. | Resultant. |  |  |  |  |
| Ether                                | $+1.6S$   | 3.15           | 4.35      | $-1.10$    |  |  |  |  |
| Toluene                              | $+0.3$  | $O^*OII$       | 0.05      | $-0.01$    |  |  |  |  |
| Carbon disulphide                    | $+0.05$   | 0.04           | 0.05      | $-0.01$    |  |  |  |  |
| Ammonia liquid<br>(con<br>centrated) | $+12$   | 1'35           | 2.01      | $-$ 0.65   |  |  |  |  |
| Benzene                              | ৰ চাতত  | აუნ            | 0.13      | $-0.06$    |  |  |  |  |
| Turpentine                           | $-0.54$   | 0.15           | 1.33      | $-0'21$    |  |  |  |  |
| Acetylene tetrachloride              | $-0.47$   | 1.67           | 2:62      | $-0.95$    |  |  |  |  |

TABLE VI (Sprayed by atomizer)

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In the case of turpentine and acetylene tetrachloride the cliarges in the substance and that in the medium are of the same sign, because the charge developed during movement through the tube of the atomizer is large compared with that developed in the breaking process. This is clearly seen from Fig. 5, which gives the charge developed in the apparatus and the substance when different quantities of turpentine are sprayed. The charge  $(x)$  of the apparatus is positive and greater than the charge  $(-x-y)$  of the substance. y is negative and has a smaller value than  $x$ .

### 7 SOLUTIONS

When distilled water is sprayed it gets positively charged, but when a small quantity of common salt is dissolved in it, it gets negatively charged. It is, therefore, of interest to consider how the positive charge changes to negative as the concentration of the dissolved substance is increased. The measurement of charge in a salt solution, when a jet strikes a jjlate was originally made by Lenard. Subsequently, Busse measured the number of ions in the air obtained with salt solution jets, but, unfortunately, did not record the charge in the substance. The experiment was, therefore, repeated. Table VII gives the charge in the substance, in the apparatus and the medium ^vhen a jet of a solution of NaCl of different concentrations is made to strike the receiver. The experiment is a difficult one, and the values of the charge in the air, particularly the low values, are to be considered as rough estimates. The results are in general agreement with those of Lenard and Busse.

### TABLE VII



## Single jet : head of water 70 cm. ; jet 8 cm. above receiver

\* Busse gave the following relative values for the total charge of possitive and negative ions in  $air =$ 



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It will be observed that the charge developed on drops changes very sharply from possitive to negative, the particular concentration at which the transition

from positive to negative charge takes place depends on the violence of the process of breaking drops. This will be clear on comparing Figs.  $6$ ,  $7$ ,  $8$ , which give the charge in the substance when solutions of NaCl of different concentration are broken into drops by the following processes :—

*ii)* By spraying, *{ii)* By making two jets strike against each other. *(iii)* By making a jet strike a plate.

Spraying by an atomizer is the most violent of the breaking processes, and transition from positive to negative charge on the liquid lakes place at a concentration of about  $\sigma'$  o25 per cent. Next to this in violence comes a single jet striking a receiver and in this case transition from positive to negative takes place at a concentration of about 0'002 per cent. Breaking of drops by making one jet strike against another is a comparatively weak process. In this case transition from positive to negative takes place at a concentration of about  $5 \times 10^{-5}$  per cent. There is apparently a reversal of the double-layer when NaCl is dissolved in water. A milder mechanical action removes considerably more positive ions than negative into the medium and thus changes the charge of the solution from positive to negative at a lower concentration than stronger mechanical action.

It will be observed from the figures given in table VII, that while the charge in the substance changes from positive to negative at a concentration of about  $\sigma$ '002%, the charge in the medium continues to remain negative even when the concentration reaches *1 %.* This is not easy to explain in view of the fact that the charge in the tube is positive and only slightly less than or very nearly equal to the charge in the substance for all concentrations from 0 005% to *1%* . The solution came out of the tube with a negative charge and this charge did not appreciably increase or decrease as a result of impact with the plate. The excess negative charge in the air may have been due to line spray from the negatively charged solution. The resultant charge as well as the ratio of negative to positive ions, however, progressively decreased as the concentration was increased to  $1\%$ . At a concentration of slightly more than  $1\%$ , the number of positive and negative ions in the air was equal When the concentration was still further increased, the number of positive ions became very slightly greater than the number of negative ions.

When a jet of dilute hydrochloric acid is broken into drops, the charge developed on the drops similarly changes from positive to negative at a certain concentration. The figures obtained in one experiment arc given below:-



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How does the reversal of the double layer take place when NaCl is added to distilled water ? Is this associated with a concentration of  $Na<sup>+</sup>$  and Cl<sup>-</sup> at the surface ? This point is discussed in § 12 *et* seq.

#### 8. SOLID PARTICLES

The case of solids has received investigation in a large number of papers by  $Shaw^{10}$  on tribo-electricity. He shows that when two identical solids, whether of ebonite, or celluloid, or silk, are rubbed together, each becomes charged in a systematic way. The charges are attributed by him to diflerential strain on the surfaces, caused by the rubbing. Lixperiments made by him in vacuum give results similar to those in open air; he, therefore, concludes that the results are entirely due to changes on the solid surfaces; aud not, even in pari, to the action of films on solid surfaces.

An effect which simulates strain; in as much as the tribo-electric state of a solid surface is changed by the process of rubbing it with a fabric, is shown by Shaw to be due to the deposition on the surface of organic films derived from the fabric. These recall the " primary " films used by Sir Wm. Hardy in researches on boundary lubrication.

It has been known that dissimilar materials when struck together in normal impact develop opposite charges.<sup>11</sup> But Shaw on trial found that identical bodies also charge one another. A glancing blow, containing tangential as well as normal stresses, is best for the purpose. The result of a violent blow between identical specimens of insulating materials is to give a net negative charge almost invariably, whether the surfaces be standard or strained. Prolonged blows in geneial increase the net negative charge.

The following results were obtained by the writer with solid particles  $(i)$ issuing as a jet through an orifice of 3 mm. and striking a receiver placed *12* cm. below, and  $(2)$  in the form of two streams moving on plains inclined at about 100'' and striking against each other in the air.

| Substance           | Weight in gm. | Single jet   | Two streams striking |  |  |
|---------------------|---------------|--------------|----------------------|--|--|
|                     | per c c, of   | charge/gm.   | charge/gm.           |  |  |
|                     | loose-powder- | e s.u        | $e, S, u$ .          |  |  |
| Aluminium (filings) | $1 - 17$      | 0.030        | 0.014                |  |  |
| Brass (filings)     | טלי ג         | $-0.034$     | $-0.336$             |  |  |
| Iron (filings)      | 2'30          | $-0.253$     | $-0.250$             |  |  |
| Dust                | 1.3           | $-0.014$     | $-3'200$             |  |  |
| Sulphur             | 1.00          | $-3.000$     | $-6.719$             |  |  |
| Chalk               | 0.58          | 0.414        | o 180                |  |  |
| Boric acid          | O' 12         | $\sim$ 21088 | $-14.558$            |  |  |
| Lead acetate        | 1.48          | O(100)       | 1.430                |  |  |

TABLE VIII

**The results are in general agreement with Shaw's observations. The impact in the air very greatly augments the charge obtained by merely making a jet strike a receiver.**

**These data may be compared with Rudge's results (qualitative) as given in table IX.**



TABLE IX

These observations show that when solid particles strike against each other **the charge they acquire is often different in .sign from that acquired by them niainly by friction through air, namely, when lliey arc sprayed, except in the case of metals.**

## PART III—SUBSTANCES INITIALLY CHARGED

## y. li) X *P* K R 1 M li N T A L DATA

**In order to determine the precise constitution of the donble-layer, cxpermients were also carried out with substances which were given initially a charge. These** show that during the process of breaking of drops, the initial charge remains **in the drops. Lach of the broken drops has a charge which is equal to its initial charge** *plus* the charge which it would get as a result of breaking if it had no **initial charge. Any** exchange **of electric charge that lakes place helueen** substance and the medium is derived entirely from the double-layer, if such a layer is supposed to exist. The initial charge, if any, remains in the substance; **it is not dislodged by the process of breaking.**

**The data for water are given in table X.**

| Distilled water-initially charged<br>TABLE X | per c.c. of water (e.s.u.).<br>Charge in air | For negative initial<br>potential.<br>For positive initial<br>potential.<br>$\omega$<br>Mean of<br>and | Resultant<br>charge.<br>Negative<br>ions<br>Positive<br>ions.<br>Resultant<br>charge.<br>Negative<br>ions.<br>Positive<br>ions.<br>$\vec{r}$ | $\overline{a}$<br>ó<br>œ<br>$\sim$<br>$\circ$<br>w,<br>+ |                | $-0.78$<br>$-1.56$<br>$4.0 + 8$<br>$-0.75$<br>$-1.56$<br>$+0.78$<br>1.58 | $\mathsf{I}$<br>ł<br>$\overline{\phantom{a}}$<br>$\overline{\phantom{a}}$<br>I<br>$\big\}$<br>1.51 | $-8.0-$<br>$-1.50$<br>$-0.0 +$<br>$-0.60$<br>$-1.32$<br>$+0.72$<br>1.54 | $-0.68$<br>$-1.36$<br>$+0.68$<br>$-0.86$<br>$-1.46$<br>$+0.60$<br>1.57 | $\mathbf{I}$<br>l<br>$\overline{\phantom{a}}$<br>$\overline{\phantom{a}}$<br>$\mathsf{I}$<br>$\overline{\phantom{a}}$<br>r·66 | ţ<br>I<br>$\mathsf I$<br>$\overline{\phantom{a}}$<br>$\overline{\phantom{a}}$<br>ł<br>$r_{53}$ | ł,                | ې<br>۱<br>$-0.095$<br>$300.0 +$<br>060.0-<br>$-0.098$<br>300.0<br>0.259 | $-0.084$<br>$-0.002$<br>$+0.008$<br>$-0.080$<br>$-0.050 -$<br>100.0<br>0.282 | I<br>I<br>I<br>$\mathbf{I}$<br>1<br>6.22.0 | ľ<br>$\overline{\phantom{a}}$<br>$\vert$<br>$\mathbf{\mathbf{I}}$<br>$\mathbf l$ |
|--|--|--|--|--|----------------|--|--|---|--|---|--|-------------------|---|--|--|--|
|  | per c.c. (e.s.u).                            |  |  |  |                |  |  |   |  |   |  |                   |   |  |  | 0.235  |
|  |  |  | For negative<br>$\begin{bmatrix} \text{initial} \\ \text{initial} \end{bmatrix}$   | ຕ  |                |  | $+0.75$  | o   | $-1.87$  | $-6.14$   | $-9.75$  |                   |   | $-1.283$   | $-2.934$                                   | $7.398$ .  |
|  | Charge in substance                          | For positive   | tial.  | $\mathbf{G}$   | Water sprayed. |  | $+2.26$  | $+3.08$   | $-5.00$  | 9,64  | $+12.84$   | .ť.<br>(b) Single |   | $+1.847$   | 45.150                                     | $+7.867$   |
|  |  | isoq) laitnətoq fatitul<br>tive or negative)<br>təloV  |  | ۳  | g              | ¢  | u  | 4   | $\overline{\mathbf{r}}$  | $\mathcal{S}^{\bullet}$   | 50   |                   | O   | $\boldsymbol{\alpha}$  | 4  | $\boldsymbol{\mathsf{s}}$  |

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 $\mathcal{L}^{\text{max}}_{\text{max}}$  ,  $\mathcal{L}^{\text{max}}_{\text{max}}$ 

 $\epsilon$ 

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It is clear from columns 2, 3 and 4 of the above table that if  $Q_0$  be the charge acquired by I c.c. of water, initially uncharged, when it is broken into drops by some mechanical process, then the same quantity of water when given an initial charge of +  $Q_r$  or  $-Q_e$  units of electricity, and broken into drops in the same manner, will acquire charge of  $Q_{\rho} + Q_{\nu}$  or  $Q_{\rho} - Q_{\nu}$ , according as the initial charge is +Q<sub>v</sub> or -Q<sub>v</sub>. This means that the initial charge of the liquid has no effect on the amount of electricity which is developed when it is broken into drops by any mechanical process. This is also confirmed by the amount of electricity in the air given in columns 5 to 10 of the above table, which remains very nearly the same whatever be the initial charge of the water.

Similar results are obtained with other liquids. Table XI gives the results obtained with acetylene tetrachloride and bromoform :-

| Potential<br>of sprayer.<br>Volts. |  | Charge per c c of acetylene tetrachloride.<br>(e, u) | Charge per c c of bromoform.<br>(e s.u.) |              |  |                   |  |
|------------------------------------|--|--|--|--------------|--|-------------------|--|
|                                    | For negative<br>For positive<br>initial poten-<br>initial poten-<br>tial.<br>tial. |  | Mean of<br>2 & 3                         | tential.     | For positive For negative<br>initial po- initial po-<br>tential. | Mean of<br>5 & 6. |  |
| I                                  | 2  | 3  | 4  | $\mathbf{5}$ | 6  |                   |  |
| $\Omega$                           |  |  | $-0.05$                                  |              |  | $-2.36$           |  |
| $\overline{a}$                     | $-0.126$   | $-1.287$   | – ს'66                                   |              |  |                   |  |
| 4                                  | $+0.536$   | $-1.854$   | $-0.00$                                  | $-0.64$      | $-3'70$  | $-2'17$           |  |
| 10                                 | $+2.808$   | $-3'168$   | $-0.08$                                  |              | $\sim$ $-$   |                   |  |
| 20                                 | $+4.482$   | $-5'901$   | $-0.71$                                  | 1.70         | $-8.71$  | $-2'27$           |  |
| 30                                 |  | $\mathbf{u}$   |  | 5.85         | $-10.95$   | $-205$            |  |

TABLE XI

#### 10 GAS JETS

Charging by a gas jet, such as escaping steam or carbon dioxide, is fairly well-known. The point of very great interest, however, is that some gas will charge the nozzle through which it escapes or the plate on which it impinges positively, while others will charge it negatively. The charge developed when a jet of gas is made to impinge on a plate depends on the nature of the gas, its pressure. the diameter of the nozzle, and the distance of the plate from the nozzle. For instance, in one experiment, carbon dioxide, issuing from a cylinder containing liquid carbon dioxide (at about 70 atmospheres) through a

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nozzle 2 mm. in diameter and striking a plate placed at a distance of 6 cm. from the nozzle, gave the following charge on the plate :—



There was vigorous ionisation, and the usual leak of i per cent, per minute of the electrometer increased to about 4 per cent, per minute in the first few minutes. The magnitude of charge decreases very rapidly with decrease of pressure and a jet issuing under very low pressure (two or three atmospheres) gives very little charge.

Acetylene, on the other hand, gave positive charge on the plate. This gas issuing under a pressure of about 18 atmospheres through a nozzle, 1 mm. in diameter, and striking a plate placed at a distance of 4 cm. would charge the plate at a rale of o'og e.s.u./second. In this case also there is vigorous ionisation and the usual leak of about i per cent, per minute of the electrometer was found to increase to about 8 per cent, per minute in the first few minutes.

Is the charging produced by an escaping gas due to the impinging of solid particles ? In the case of escaping steam, we liavc the water drops impinging on the walls of the nozzle The CO<sub>2</sub> comes out from a cylinder of liquid carbon dioxide probably as solid particles. Acetylene gas is not usually compressed into cylinders but is dissolved in acetone, with which the porous mass inside the cylinder is impregnated, and, therefore, when it escapes, it probably carries with it a fine spray of the liquid. We have, however, the following statement by C.T.R. Wilson<sup>12</sup> :— "According to a large class of theories the air in contact with the ground acquires a positive charge, *i.e.*, by friction." Are we to assume that there is a double-layer between air and earth's surface, and that the movement of air causes a slip, there being no appreciable separation of charge unless the velocity is great ? •

If we assume that there is an electrical double-layer in operation, it cannot he that the outer charge of the normal double-layer between the plate and air is removed by the impinging gas, for in that case the plate would show charge of the same sign both in the case of carbon dioxide and acetylene. Can the development be explained by the following two processes :—

( $I$ ) At the nozzle, there is a double-layer between the gas and the metal surface of the nozzle, and the frictional action on this double layer gives the issuing gas a charge.

(2) When this charged gas impinges on the plate a similar double-layer is instantly formed between the gas and the plate, and friction accentuates the charge on the already charged gas.

## II. MECHANICAL ACTION IN THE ATMOSPHERE

In the atmosphere we get numerous examples of generation of charge by mechanical action. Over the sea surface, the splashing of sea water makes the water negatively charged and this undoubtedly plays a very important rôle in the replenishment of the earth's negative charge. In thunderstorms, we have examples of water drops striking against each other or being broken in a vertical blast of air. There are also the ice particles striking against each other and getting negatively charged in the higher region of the clouds. The generation and distribution of charge in thunderclouds are subjects which have received elaborate investigation in recent years. In sandstorms and snowstorms (blizzards) we have other examples of generation of charge by mechanical action. Fog particles probably get charged by condensation on ions or charged nuclei or by a slip of the double-layer during movements through air.

It is essential to realise an important distinction between the processes in operation in the atmosphere and those in the laboratory. In the laboratory we are unable to generate charge without making use of some sort of apparatus, and the reaction between the apparatus and the liquid or solid particles always makes the drops or the solid particles leave the apparatus with an initial charge. Earthing the apparatus makes no difference; while the charge in the apparatus escapes into the earth, the drops . or the solid particles leave it with an equal and opposite charge. In these experiments the total charge is shared between the apparatus, substance, and the medium. The initial charge profoundly affects the relationship between the charge in the substance and that in the air, and we sometimes find that they are of the same sign. For instance, when a solution of NaCl is broken by spraying or by making a jet strike a plate, the charges in the substance and the air are of the same sign for the range of concentration  $\sigma$  005% to  $1\%$ . When'drops are broken in a vertical blast of air, they come out of the tube with an initial charge, with the result that while the drops after being broken by the air current get a charge of  $+o'$  o23 c. s. u./c.c.) the total charge in the air is less than half this amount. In the atmosphere, there is no apparatus; the drops may form on ions or nuclei, and may have an initial positive or negative charge, but if they are broken in a current of air, the total charge in the air which will be derived from I c.c.

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will be roughly  $-o'$  coog2 c,s.u. and the charge acquired by the drops by this operation will also be equal to this amount but positive in sign. This *plus* the initial charge will determine the total charge of the drops. If the drops break by striking against each other, the charge in the air will be roughly $-$ o'o6o c. s. u./c.c. and in the drops the initial charge  $+$ o  $\infty$  e. s. u, per c.c.

We may give here one example showing how the calculation of charge in atmospheric processes is modified when we take these aspects into consideration. As explained in a previous paragraph, with, increasing amounts of dissolved salts (NaCl) or acids (HCl) in distilled water, the charge developed on drops changes very sharply from positive to negative. It also appears from these experiments that when drops are broken by a comparatively mild process, such as, by making one jet strike against another or in a vertical blast of air, the transition from positive to negative takes place at a very low concentration, roughly of the order of  $5 \times 10^{-5}$  per cent.

Now, it is known that when evaporated to dryness, 100,000 parts by weight of rain water will give about  $\sigma$ '34 part of solid matter and that most of this consists of sodium chloride and organic matter. This is equivalent to a concentration of  $3'4 \times 10^{-3}$  per cent. Near sea coast and towns the impurities are usually more than this amount. As the impurities are collected during the journey through air, the contamination may be regarded to be slightly less than this amount in the region where drops are first formed; also the rain which falls at the end of a shower is less contaminated than that which falls at the beginning. We shall, therefore, not be far out if we assume that the concentration of chlorides in rain drops is always greater than  $10^{-4}$  per cent, except towards the end of a shower. If, therefore, charge is developed in a thundercloud by breaking of drops or by drops striking against each other, and if the salt content of rain drops be as much as the average value ( $\sigma$ ' $\cos 34\%$ ) or more, the charge developed is either very small positive or negative. We are so near the transition point that *we* can develop very little charge by breaking of drops. It is only when the drops have very little contamination that wc can develop a large quantity of positive charge by breaking them. Simpson's calculation of sparking potential in clouds produced by the breaking process would, therefore, seem to need a revision. It is also probably due to this reason that the breaking process is no longer found to play the same important part in the generation of charge in thunderclouds as it was once supposed to do.

#### 12. HOW IS CHARGE DEVELOPED?

Wc now proceed to consider the different factors which may contribute, to the generation of charge in the various processes employed.

We know that when crystals are split, charges<sup>13</sup> are found, for instance in calcite, selenite, mica, fluorspar, gypsum, etc. Is the charge developed by strain, disruption, cleavage, deformation, etc., a pure consequence of removal of electrons from or addition of electrons to the two surfaces by the forces in operation ? As has been already pointed out by Shaw, the following factors come into consideration in this connection :—

(1) The contact effects<sup>14</sup> somewhat akin to the Peltier P.D, between metals.

(2) The rôle of absorbed films.  $15$  According to Nernst's theory the metals go into solution when brought into contact with liquid films, each metal having its own solution pressure and receiving a corresponding greater or less negative charge. Coehn supposes that even insulators like glass are merely bad electrolytes, so that even for these solid contacts the Nernst principle holds good. When a liquid rubs a solid we should have the electrokinetic phenomenon of "stream potential," the converse of electro-osmose, whereby the solid would be charged according to Helmholtz's formula. It has been shown by various writers, notably Knoblauch, that an acid surface has itself a negative, and an alkaline one a positive tendency. In the first case the surface loses  $+ (H)$ ions in the latter  $-(OH)$  ions. As surfaces are commonly acidic or alkaline, this must be an important factor in the generation of charge.

(3) According to the electron theory of matter the interface P.D. varies as the absolute temperature, and, consequently, the temperature changes caused by friction, disruption, fusion, evaporation, etc-, must contribute to the production of charge.

( $_4$ ) The physical changes in the surfaces as a result of rubbing, etc, and their rclationship to the tribo-electric properties constitute another very important factor according to Shaw.

Therefore, the net potential V can be expressed as

$$
V = E - P \pm S - M \pm A - T,
$$

where  $E^{\text{in}} = E^{\text{in}}$  Electron surrender causing a P.D. akin to the Peltier P.D. P = solution pressures (Nernst), S = strain effect (Shaw),  $M =$ moisture effect (French, Vieweg),  $A = acid effect (positive)$ (Knoblauch), alkaline effect (negative) T = efl'cct due to temperature changes at the interface.

In any specific case, one or the other factor may be absent, and some factors may become more important than the others.

Coming to the question of electrical double-layer, Ficnke. interesting paper on the surface electrical double-layer of solid and liquid bodies showed mathematically that if the atoms be assumed to consist of positive nuclei with electrons rotating about them, these electrons constitute over the surface of the metals a layer of negative electrification with a corresponding layer of positive electrification on the inner side of the surface, the electric dcuble-layer of atomic thickness thus formed, preventing the free electrons from escaping. If, in the

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same way, a double-layer is supposed to be formed on the surface of any solid, liquid or gas, then at an inter-face between two dissimilar substances, the charge **on either face will be the net result of the positive charge on its inner side** *plus* the negative charge on the upper side of the other face. If, in any case, the net effect of the combination on each of the two faces in contact happens to be a **zero charge then clearly there is no double-layer formation. This will be the case at the interface between two substances of the same material. This may explain** why by breaking a liquid in its own vapour we can develop very little charge.

There is thus evidence regarding the formation of a double-layer on the surface of all uncharged substances. As regards charged substances, we have **seen that, in the exchange of electricity between a liquid and the surrounding** medium, when the former is broken into drops, the initial charge of the liquid does not come into play. The initial charge of the substance must, therefore, be considered to be adhering to it during the process of division. The doublelayer, therefore, must be considered to be a natural formation at the boundary between two surfaces in contact, between solid and solid, solid and liquid, solid and gas, liquid and gas, etc., which remains inactive until there is a slipping or disruption; it is formed whether the substances are initially charged or uncharged. It is purely a boundary phenomenon and all exchanges of electricity during the processes described, takes place from this boundary phenomenon. In any case, the assumption of the existence of an electrical doublelayer enables us to explain the observed effects more easily than on any other assumption.

While we cannot say we understand yet fully how the charges are developed we must say that we have gained in recent years a good deal of insight into the processes in operation.

#### **R K F** *n R* **K N C B vS**

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**16** *Slaiistical Mechanics* **(Second Kdition), Chapter X lII, application to liquids and solutions, also p. 824,** *et seq* **discussing Langmuir's work on the subject**

*Phil. Mag.* 33**, 297 (1917),**