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CONDENSATION NUCLEI.


If we take the ordinary air of a room, and enclose it in a glass vessel containing some water and provided with some means of increasing or diminishing the volume at will, we are able to observe the following phenomena. If the air has been allowed to stand sufficiently long to become saturated with water vapor, any increase of volume, even if very slight, causes the formation of a fog throughout the volume of the moist air. This is easily made visible by concentrating a powerful beam of light on the contents of the vessel; or, by placing a small source of light behind the vessel, brilliant colored rings or coronas may be seen surrounding the source. If the air be made to contract again to its original volume, a second expansion like the first will again give a similar fog, but when this process has been several times repeated, the fogs become thinner, the drops being fewer and larger; we get at length a fine rain on expansion rather than a fog, the drops falling to the bottom of the vessel within a few seconds, instead of remaining in suspension for many minutes like the first-formed fog particles. When this stage has been reached, the next and all succeeding expansions produce no drops at all, no condensation resulting elsewhere than on the walls of the vessel. If ordinary air be now admitted into the vessel, drops will again be seen on expansion, unless the air introduced has entered through a tightly pressed plug of cotton-wool, or has been otherwise filtered, in which case no drops are seen.

The phenomena are readily explained if we suppose that water cannot under ordinary circumstances condense in the form of drops unless suitable nuclei are present to serve as starting points for the drops. These nuclei are present in very varying numbers in ordinary atmospheric air, from which they may be removed by filtering, or by repeatedly forming a cloud by expansion, and allowing the drops to fall to the bottom of the vessel. Both the facts and
the explanation have been long known. The particles which serve as the nuclei of the drops formed, when ordinary atmospheric air is allowed to expand slightly, are conveniently called "dust" particles; they are generally too small to be themselves visible, and it would be difficult to find a means of determining whether they consist of solid particles or of minute drops of liquid. The number of these dust particles per c.c. of air in different localities and under different weather conditions has been investigated by Aitken, and by others, with the aid of his ingenious dust-counting apparatus.

It is not difficult to understand why nuclei should be necessary for the condensation of water in the form of drops. Lord Kelvin proved that the pressure of aqueous vapor necessary for equilibrium over a convex or concave surface of water differed from that over a flat surface, being less over a concave and greater over a convex surface. He shows how we may calculate the difference. A very small drop of pure water will, if we assume the surface tension to remain the same for very small drops as for large ones, evaporate even when surrounded by vapor many times more dense than that in equilibrium at the same temperature over a flat surface. Thus unless the initial stages of the growth of the drops can be, as it were, omitted, owing to the presence of not too minute nuclei, a high degree of supersaturation may exist without any condensation in the form of rain or cloud resulting. Lord Kelvin showed that to alter the equilibrium vapor-pressure by one part in a thousand, the radius of curvature of a spherical drop must amount to about $10^{-4}$ cm. Thus very minute nuclei will enable a cloud to be formed with a very slight degree of supersaturation, in other words, as a result of a very slight expansion of the air if this has been initially saturated with water vapor.

Lord Kelvin refrained from extending his calculations to curvatures of greater amount, as the surface tension cannot remain independent of the radius much beyond that limit. It is convenient, however, to extend the calculations to greater curvatures; for although the results obtained cannot be considered as quantitatively correct, they enable us to form a picture of the mode of action of nuclei of different kinds. Let us imagine an arrangement equivalent to that considered by Lord Kelvin1; but since we are

here concerned with convex surfaces, let the capillary tube be joined as a side tube to the lower part of a tall vessel of water. The capillary must be supposed to have walls of such a nature as not to be wetted by water, and let us suppose the open end of it to be bent round, so that it points vertically upwards, and that the height of the vertical portion can be adjusted to bring the meniscus to the open end of the tube. Let the whole apparatus be contained in a closed vessel containing only water vapor.

We have then the convex water-air meniscus depressed below the level of the flat surface in the large vessel to a depth \( h \), such that \( gh h = \frac{2T}{r} \), where \( g \) is the acceleration due to gravity, \( w \) the density of the liquid* (\( \omega = 1 \) in the present case), \( T \) is the surface tension and \( r \) the radius of curvature. Thus the pressure of the vapor in contact with the meniscus must be greater than that over the flat surface by that due to the weight of a column of water vapor of height \( h \), the pressure at the top of the column being that required for equilibrium over a flat surface at the given temperature. This increased pressure must, moreover, be the pressure necessary for equilibrium over the curved surface, distillation from the one surface to the other would otherwise take place resulting in a continuous circulation. To find this pressure \( p_2, p_1 \) being that at the flat surface, we have \( dp = g \rho \, dh \),

\[
h = \frac{1}{g} \int \frac{dp}{\rho}
\]

\( \rho \) being the density of the steam. If we assume Boyle's law to be obeyed, this gives

\[
h = \frac{R}{g} t \log_\rho \frac{p_2}{p_1} = \frac{R}{g} t \log_\rho \rho_2
\]

\( R \) being the constant in the equation \( re = Rt, t \) being the absolute temperature, \( \rho_1, \rho_2 \) the density of the vapor at the two surfaces respectively. But \( h = 2T/rg \), thus

\[
\log_\rho \frac{p_2}{p_1} = \log_\rho \frac{\rho_2}{\rho_1} = \frac{1}{R t} \cdot \frac{2T}{r}.
\]

We have thus the means of calculating the pressure, or the density, which water vapor must have, in order that it may be in equilibrium in contact with a drop of any size. The equilibrium is obviously unstable, a drop if too big for equilibrium will grow,

* The weight of a column of the vapor is neglected in comparison with that of a column of the liquid of the same height.
so long as the supersaturated condition is maintained, if too small it will evaporate completely. The possession of a charge of electricity by the drop, or the existence of a dissolved substance within it, will cause the drop to be stable, if its size be less than a certain limit, depending on the magnitude of the charge, or the quantity of dissolved substance. Let us consider the case of electrification. We may imagine the water surface in one limb of a U-tube, in an arrangement like that described above, to be uniformly charged with electricity, by holding a very short distance above it a parallel conducting surface maintained at a different potential. It is immaterial whether the water surface be flat or curved; a tension of $2 \pi e^2$ dynes per square cm will be exerted on the end of the column, $\sigma$ being the charge per sq. cm. This will raise the electrified surface, above the level which it would have occupied in the absence of the charge, through a distance $2 \pi \sigma^2/g$ and there will be a corresponding diminution in the saturation vapor-pressure. The vapor pressure necessary for equilibrium over a charged drop is now given by the equation

$$\log_e \frac{p_2}{p_1} = \frac{1}{Rt} \left( \frac{2T}{r} - 2 \pi e^2 \right)$$

$$= \frac{1}{Rt} \left( \frac{2T}{r} - \frac{e^2}{8 \pi r^4} \right)$$

where $p_1$ is the saturation vapor-pressure over a flat uncharged surface, $p_2$ that necessary for equilibrium at the same temperature in presence of the drops, and $e$ is the charge on each drop. In an atmosphere saturated with respect to a flat uncharged surface, a drop carrying a charge $e$, would be in stable equilibrium if its radius were such that the two terms on the right-hand side of the above equation were equal, i. e., when $r^2 = e^2/16 \pi T$. If the density of the vapor were increased, the drop would become larger, the equilibrium remaining stable until the vapor-pressure reached the maximum value corresponding to the above equation. To find this we have on differentiating

$$\frac{1}{\rho} \frac{dp}{dr} = \frac{1}{Rt} \left( -\frac{2T}{r^2} + \frac{1}{2} \frac{e^2}{\pi r^5} \right)$$

The maximum vapor-pressure in contact with the drops occurs when $r^2 = e^2/4 \pi T$, and has the value given by

$$\log \frac{p_2}{p_1} = \frac{3T}{2Kt}$$
If the pressure of the vapor be increased beyond this limit, the unstable condition is reached, and the drop increases in size so long as the supply of vapor is unlimited. In most cases the final size of the drops would be determined by the amount of vapor initially present, and the number of drops among which the water is distributed; unless they are very numerous, and, therefore, very small when full grown, they will grow until the vapor is not sensibly supersaturated; it will only be in very rare cases that the final size of the drops is so small that equilibrium will be reached while the vapor is at all supersaturated.

It is easily seen that the behavior of drops containing dissolved substances will be quite similar; if we start with very small drops, there is for a given size of drops a certain vapor-pressure corresponding to equilibrium, if we increase the density of the vapor the drop grows, the equilibrium remaining stable, until a certain size is reached, after which the drops suddenly grow to their full size. The theory of condensation on ions or other nuclei has been treated by J. J. Thomson and by Langevin and Bloch.

LIMITING SUPERSATURATION IN DUST-FREE GASES.

When air saturated with water vapor has been freed from dust particles, no drops are formed on expansion, provided that a certain critical degree of supersaturation has not been exceeded. To produce the supersaturation necessary for condensation in the form of drops in dust-free air, the air must be allowed to expand suddenly, till the final volume is 1.25 times the initial volume. The condensation is rainlike in form, and the number of drops remains small although the expansion considerably exceeds this lower limit. Expansions exceeding a second limit, $v_2/v_1 = 1.38$ give fogs, which increase rapidly in density, i.e., in the number of the drops, as the expansion is increased beyond this limit. In such experiments it is of course necessary that the apparatus used should be such that a very rapid change of volume can be brought about, and that the ratio of the final to the initial volume is known with certainty. Some years ago I introduced a method which has proved suitable for the purpose. When this method is

4. C. T. R. Wilson, Phil. Trans. 189, p. 265, 1897.
used, it would appear from the consistency of the results obtained with cloud-chambers varying in capacity from 15 to 1500 c.c., that the expansion is adiabatic and is completed before any appreciable quantity of water has had time to separate out. From the ratio of the final to the initial volume, knowing the initial temperature, we can deduce the temperature at the moment when the expansion was completed from the equation for the cooling of a gas by adiabatic expansion,

\[ \frac{\theta_2}{\theta_1} = \left( \frac{v_1}{v_2} \right)^{\gamma-1} \]

\( \gamma \) may be taken as not differing sensibly from its value for the dry gas. Knowing the final temperature we have the data from which we can obtain the density of the vapor which would be required for saturation at the moment of completion of the expansion, and we know the actual density at that moment from the initial temperature and the ratio of the final to the initial volume. Thus the supersaturation, measured by the ratio of the actual density of the vapor at the instant when the expansion is completed, to the density of the saturated vapor at the temperature which the supersaturated gas then possesses, can be calculated.

The supersaturation required for the rainlike condensation is found in this way to be approximately fourfold, that required for the cloudlike condensation being nearly eightfold. There are these two classes of nuclei always present in moist dust-free air, and always being produced, for, however often the process of condensing water on the nuclei and allowing the drops to settle is repeated, the number of drops formed in subsequent expansions is undiminished. The nuclei which give rise to the rainlike condensation and which are at any moment present in quite small numbers are, as we shall see, ions continually being produced in the gas. They can be removed by an electric field. The cloudlike condensation occurring with large expansions is entirely unaffected by an electric field; it is independent also of the nature of the gas. If we calculate how large a drop of water would require to be in order that it should just be able to grow in vapor of eightfold supersaturation, we obtain the very small value \( 6.4 \times 10^{-8} \) cm for the radius of such drops. Thus drops not large in comparison with molecular dimensions might be expected to grow into visible drops in an atmosphere supersaturated to this extent.
THE IONS AS CONDENSATION NUCLEI.

If we expose the cloud chamber of an expansion apparatus to the action of Röntgen rays, the air having been previously freed from dust, just the same expansion is required as in the absence of the rays to produce drops, but now we get comparatively dense fogs in place of the rainlike condensation. The cloudlike condensation obtained with expansions exceeding the second limit is not sensibly affected. Thus, when X-rays pass through moist air, they produce nuclei of exactly the same efficiency in promoting condensation, as those which are always being produced in small numbers, and to which the rainlike condensation is due. The conducting power imparted to air by the action of X-rays being explained as due to the setting free of ions in the gas, it was natural to identify the nuclei with the ions.

This view was verified by studying the action of an electric field on the nuclei produced by X-rays. Between two parallel plates, which formed the top and bottom of the cloud chamber of an expansion apparatus, a difference of potential of some hundred volts could be applied. With the electric field acting the number of drops produced on expansion in air exposed to the rays was exceedingly small in comparison with the number seen in the absence of the field. The nuclei carry a charge of electricity, and are driven by the electric field against the plates immediately after being set free. The direct proof that the few nuclei, which are always present and which give rise to the rainlike condensation, are also ions has been more difficult to carry out. Attempts made with small apparatus led to negative results, the number of drops being inconveniently small whether the field was applied or not. Recent experiments on a large scale, however, showed in a striking way the removal of these nuclei by the electric field. The subject has been further cleared up by proof by purely electrical measurements that the air in a closed vessel is continually being ionised.

Air ionised by any of the various types of Becquerel rays, or containing ions from a zinc plate exposed to weak ultra-violet light, behaves, on expansion, like air exposed to X-rays; fogs being produced in air initially saturated if the lower expansion limit \( v_2/v_1 = 1.25 \) be exceeded. The action of an electric field in re-

moving the nuclei is the same in air ionised by Becquerel rays, as in air ionised by X-rays. The ions produced by the discharge from a point are similar in their action; but there is here a tendency, due probably to the products of chemical combinations brought about by the luminous discharge, for the nuclei to grow, or for larger uncharged nuclei to be formed, so that a much smaller degree of supersaturation may be required to produce a cloud. The ions produced by these various methods are also identical in the velocity with which they move through air under a given potential gradient. The degree of supersaturation required to make water condense on the ions is independent of the gas.

If we make use of the equation which has been given above, connecting the maximum supersaturation with the charge of the drop, we obtain the result $e = 6 \times 10^{-10}$ electrostatic units for a fourfold supersaturation. To obtain this number we have of course extended to drops of almost molecular smallness, \( r = 7 \times 10^{-8} \, \text{cm} \), an equation which could only be used with confidence when the radius was at least a thousand times as great. It is, therefore, somewhat remarkable that the value obtained approximates fairly closely to the values found by J. J. Thomson and by H. A. Wilson for the ionic charge. The action of the ions as condensation nuclei is not, however, completely explained, for our formula would make efficiency of the electrification in helping condensation independent of the sign of the charge. Now the negative ions are found to require a less degree of supersaturation to make water form visible drops upon them than do the positive.

**Difference between Positive and Negative Ions.**

To study this question, we may use an expansion apparatus provided with a cloud chamber, in which the air under examination is contained between two horizontal plates kept at slightly different potentials. A thin stratum of the air immediately over the lower plate is exposed to the action of X-rays. A series of observations are then made in which the rays are cut off at a definite interval of time before the expansion is made, the interval being such that all the downward moving ions have had time to reach the lower plate, while only a small proportion of the upward moving ones have reached the much more distant upper plate before the

expansion takes place. Thus, at the moment of expansion, we will have practically ions of only one kind present, those namely which are moving toward the upper plate.

In this way it has been found that in order that water may condense upon them to form visible drops, the negative ions require an expansion $v_2/v_1=1.25$, the positive an expansion 1.31, the corresponding supersaturations being fourfold and sixfold respectively.

When ions of both kinds are present in approximately equal numbers, it is often possible to observe a difference in the density of the resulting cloud according as the expansion is below or above the limit corresponding to the degree of supersaturation necessary for the condensation of the positive ions. The increase in density was first described by J. J. Thomson, and it was suggested by him that it might be due to a difference between the positive and negative ions in their efficiency as condensation nuclei; he pointed out that such a difference, if established, would have important bearings on the subject of atmospheric electricity. For an electrical field might be expected to result in ionised air if such a degree of supersaturation was reached that condensation took place on ions of one kind only, these loaded ions being then carried down by gravity. That the drops produced under these conditions are actually negatively charged, as was to be expected from the greater efficiency of the negative ion as a nucleus, was proved by H. A. Wilson, by observing the movement of the drops in a strong electric field applied after their formation by expansion.

Charge Carried by the Ions.

The most important use which has been made of the fact that ions act as nuclei for the condensation of water vapor has been in the determination of the quantity of electricity carried by each ion. Two entirely different methods have been employed, both requiring the use of the expansion apparatus. In the first, that of J. J. Thomson, a measurement of the leakage of electricity through the air of the cloud chamber allows $n\cdot e$, the product of the number of ions and the ionic charge, to be measured; $n$, the number of the ions is given by the number of the drops. The number has been obtained, not by direct counting, but from a knowledge of the quantity of water condensed, and the size of the drops

8. J. J. Thomson, Phil. Mag. v. 46, p. 528, 1898; Phil. Mag. v. 48, p. 547, 1899; Conduction of Electricity through Gases, p. 121.
as obtained from their rate of fall. The second method (used by H. A. Wilson*) in its simplest form reduces itself to a determination of the strength of the electric field necessary to maintain in suspension the drops condensed upon the ions. We then have $F' e$, the product of the strength of the field and the charge on the drop, equal to its weight. The size of the drops, and hence their weight, is again deduced from the rate of fall in the absence of the field.

**Other Properties of the Ions.**

There is no room for doubt that the nuclei produced by X-rays and similar agents, and requiring a fourfold or sixfold supersaturation to make water condense on them, are negatively or positively charged ions. We know by other methods of studying them a great deal about the properties of ions, their velocity in an electric field, their diffusion constants and rates of recombination under different conditions. Their behavior when studied by condensation has been entirely in agreement with the results obtained by other methods; for example, the rapidity with which their number diminishes after the source of ionisation has been cut off.

**Nuclei Similar in Efficiency to the Ions, but not Removable by an Electrical Field.**

Moist air exposed to weak ultra-violet light is found to contain a plentiful supply of nuclei, which require a degree of supersaturation approximately the same as do the ions, in order that a cloud may form upon them. Yet even very strong electric fields appear to be without effect in reducing the number of drops formed on expansion. Certain metals also produce in the air in contact with them similar nuclei, the clouds in this case, however, not generally attaining any considerable density, unless the expansion is great enough to cause condensation on positive ions. It is possible that we have in both these cases ions produced as a result of the expansion, there being, therefore, no time for the ions to be removed by the field before the cloud is formed.

**Nuclei More Effective in Promoting Condensation than the Ions Produced by X-rays.**

If we expose moist air to ultra-violet of moderate intensity, the result is not so simple as when the intensity is very small. Nu-

nuclei are produced, which appear to grow under the action of the light, the expansion required to produce a cloud becoming less than that required by the negative ions, and becoming less and less the stronger the light and the longer the exposure. For a given intensity of the light, there appears to be a maximum size beyond which the nuclei cease to grow. A very moderate intensity is sufficient to produce nuclei which grow till the slightest expansion will form a cloud, and the growth is very rapid so that the earlier stages are difficult to follow. With very intense ultra-violet light, the growth continues till the nuclei become visible in suitable illumination, and we get a cloud without expansion, even in unsaturated air. There can be little doubt that the growth of these nuclei into visible drops is to be attributed to the formation of some substance in solution within them. Vincent has recently studied these visible nuclei and found some of the particles to be positively, some negatively charged, and others neutral; but he finds the evidence to be in favor of the view that the charges are, as if were, accidental, being simply due to ions which have come in contact with them. Lenard had previously shown the ionisation of the air by these rays.

The very small nuclei, i. e., those which require large expansions to make drops form upon them, diffuse rapidly to the sides of the vessel, so that a fog is not formed if the radiation be cut off even one minute before the expansion is made; the nuclei which are large enough to be visible may persist for hours on account of their very slow diffusion.

Other nuclei, which like those produced by ultra-violet light, vary in size with varying conditions, are those produced by heating a wire, studied some time ago by Aitken and recently by Owen. The latter has shown that the lower the temperature at which they have been given off by the wire, the greater is the expansion required to catch them. They can be detected when the wire has been raised to a temperature of less than 150° C. in air. The nuclei produced by the slow oxidation of phosphorus, like those formed by the action of strong ultra-violet light, form visible clouds in air which is not supersaturated. These clouds have been studied by Barus and others. As in the cases just considered, the production of the nuclei is associated with the acquisition of conducting power by the gas. There has been a considerable amount of controversy as to the nature of the conduction of elec-
tricity in air which has passed over phosphorus. The experiments of Bloch have, however, proved from the nature of the curve obtained for the relation between current and potential difference, that we have here a true case of ionisation. His measurements of the velocity of the ions showed that they have a very small mobility as compared with the ions due to X-rays. His experiments leave little room for doubt that these slow-moving ions are identical with the nuclei. The mobility is about a thousand times smaller than that of the ions formed by X-rays.

Certain experiments of Harms, and of Elster and Geitel, appear to show that by the oxidation of phosphorus, free ions are produced, in addition to the visible cloud particles. These we should expect to be rapidly removed by diffusion and recombination, and, after passing through any considerable length of tubing, we should expect only the loaded ions to persist. The absence of unloaded ions in Bloch's experiments is perhaps to be explained in this way.

The nuclei found in freshly prepared gases, and studied especially by Townsend, resemble in many ways those resulting from the oxidation of phosphorus. Like them, they form clouds without supersaturation, and they carry a charge of electricity. In some cases at least, as was shown by Townsend's experiments, the charge on each nucleus is the ionic charge. Bloch has studied the mobility of these ions, and in agreement with Townsend has found it to be of the same order as that of the phosphorus ions.

By the splashing of water or aqueous solutions, or the bubbling of air through water or solutions, nuclei are produced requiring only a slight expansion in order that water may condense upon them. These nuclei have lately been studied by Barus. He finds that the nuclei produced from salt solutions are much more persistent than those arising from distilled water. It is most natural to regard these nuclei, as does Barus, as small drops which have evaporated till the strength of the solution is such, that the effect of the dissolved substance on the vapor-pressure counterbalances that of the surface tension. The splashing or bubbling process also imparts temporary conducting power to the gas. According

to Kaehler\textsuperscript{14}, with pure distilled water the conduction is practically unipolar, and due to the presence of negative ions having a mobility equal to that of the ions produced by X-rays; with salt water positive ions of very small mobility are produced in addition.

In the products of combustion from flames, we find again ions of small mobility, and no appreciable degree of supersaturation is required to produce a cloud.

As Bloch points out, there are apparently two classes of ions. We have first ions like those produced by X-rays and similar agents, which have a definite velocity in an electric field of given strength, and require a definite degree of supersaturation, fourfold or sixfold according to the sign of the charge, in order that water may condense upon them. The second class consists of ions of variable mobility, about one-thousand part of that of the ions of the first class, and they have the power of condensing water to form visible drops without supersaturation. Ions with intermediate properties are rarely, if ever, met with. Bloch points out that we should expect an important difference between the two classes with respect to the result of recombination of positive and negative ions. In the first class the nucleus owes its existence to the charge; if two oppositely charged ions (which we may regard as minute charged drops) combine, we should expect the resultant uncharged nucleus to evaporate at once. On the other hand, the persistence of the ions of the second class cannot be due to the charge alone, and neutralisation of the charge will not result in evaporation of the nucleus. From recombination of these ions persistent uncharged nuclei will result. The facts are found to be in complete accord with these considerations.

If we produce a cloud in dust-free air by an expansion exceeding 1.25 after exposure to X-rays, or exceeding 1.38 in the absence of ionising agents, the drops formed, if made to evaporate by compression, appear to leave behind nuclei requiring only slight expansion to make water condense on them. J. J. Thomson has pointed out that there may be a certain size for which even uncharged drops of pure water may be stable in an unsaturated atmosphere. For, according to the experiments of Reinold and Rücker, the surface tension of thin films has a minimum for a certain thickness. There may, therefore, be a certain size (somewhat

\textsuperscript{14.} Kaehler, Ann. der Phys. XII, p. 1119, 1903.
smaller than that corresponding to minimum surface tension) for which the potential energy of a drop due to surface tension has a minimum value. Such a drop would be in equilibrium in vapor saturated with respect to a flat surface.

Bloch, following Langevin, works out, in the paper already referred to, the theory of condensation of water vapor on ions. He shows that we might expect drops of about 10 μ in diameter to be stable, on account of the variation in surface tension in that region, but we should not expect to meet with drops of which the diameter was comprised between that limit and a very low value, the equilibrium of such particles being unstable. The behavior of other substances than water would probably be similar. In this way Bloch explains the fact that we do not meet with ions of mobility intermediate between about 1 cm and 1/300 cm per second for a field of 1 volt per cm.

There are then three principal classes of nuclei: (1) The ions proper, requiring a fourfold or sixfold supersaturation to cause water to condense on them, and having a mobility exceeding 1 cm per second in a field of 1 volt per cm; (2) loaded ions requiring little or no supersaturation to make water condense on them, and having a mobility generally less than a thousandth part of that of the ions proper; (3) uncharged nuclei, resembling the second class in requiring little or no supersaturation in order that visible drops may form upon them.