

THE BEHAVIOR OF CONDENSATION NUCLEI UNDER CHANGING HUMIDITIES

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Natural condensation nuclei range from about $10\ \mu$ to less than $0.01\ \mu$ in radius. Above $1\ \mu$ radius, nuclei size and humidity relationships follow, in general, the theory of Köhler [1] except that the points of solution do not exactly occur at relative humidities predicted from the properties of the bulk solution. Disagreement is most pronounced with the smaller sizes as shown by Junge [2]. To what extent this behavior carried into the size range of the more numerous Aitken-nuclei, those with radii between 0.01 and $0.1\ \mu$, was not known. The question is particularly important with regard to the supersaturation at which nuclei become active for condensation. Since measurements of supersaturation are difficult to perform and since results can be obtained by extrapolating data on size change with humidity if the extrapolation is verified, a thorough study of the growth of Aitken-nuclei of known composition with humidity was undertaken.

A diagram of the apparatus is shown in fig. 1. As may be seen it consisted essentially of: 1. air cleaning and pressure regulating equipment; 2. an ion or aerosol generator; 3. devices for sharpening the ion size distribution; 4. controlled humidity chambers; and 5. the ion counter with its attached electrical equipment.

Ion generation, except in the case of insoluble AgI, was accomplished by atomizing dilute solutions of the compounds to be studied with dry air. A DeVilbiss No. 180 atomizer, using 7.1 lit per min of air at 13 lb per in² pressure and having an impaction plate set in the aerosol stream to cut out droplets larger than about $1\ \mu$, was used. The atomizer was maintained at ice temperature to reduce the quantity of moisture evaporated. After leaving the generator, the aerosol

stream bubbled through 30 in of water in a 3-in vertical column and then passed through a device consisting of a 0.6-in inside diameter stainless steel cylinder 6.5 in long with a $\frac{3}{16}$ -in outside diameter coaxial inner electrode and having a 45-v potential difference between the inner and outer conductors. The bubbler preferentially removed the large particles and the second device preferentially removed the smaller ions. The combination resulted in an aerosol or ion stream having a rather sharp size distribution wholly within the desired size range. Being quite insoluble, ions of AgI were obtained by vaporizing the compound from an electric furnace into a flowing nitrogen stream, following essentially the method of Manson [3].

The conditioning chambers and the aging chamber served to establish equilibrium conditions and to determine the direction from which equilibrium was attained by the aerosol particles. A high temperature in the conditioning chambers resulted in a relative humidity of less than 15 per cent. Upon flowing from the conditioning chambers at an elevated temperature to the aging chamber at ambient temperature, the

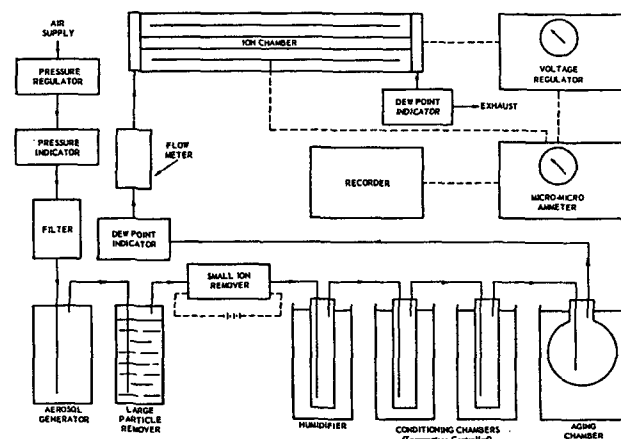


FIG. 1. Schematic diagram of apparatus.

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humidity increased and the resulting state of the ions was that attained in going from this low relative humidity to a higher relative humidity. Conversely, with the conditioning chambers at a low temperature and the aging chamber at the ambient temperature the aerosol state resulting was that attained in going from a relative humidity above 85 per cent to a lower humidity. By bringing about shifts in relative humidity through temperature change all nuclei were subjected to the same humidity conditions. Had addition or extraction of moisture been employed instead, differences in nuclei sizes might have introduced spurious effects and sharp transitions at the solution or crystallization points might not have been obtained.

The ion chamber consisted of two coaxial electrodes in an electrostatic shield insulated from each other and from the shield. The inner and outer electrodes were maintained at an adjustable, known potential difference. The outer electrode was negative, and the current flowing to it, the positive ion current, was measured. Electrical insulation was provided by teflon which was maintained by means of heaters some 5 to 10C above other parts of the counter to prevent the possibility of short-circuiting by condensation. The inlet and outlet ends of the ion chamber were arranged with double plenum chambers designed to distribute the air flow uniformly and to insure uniform laminar flow between the electrodes. The flow pattern for the flow rates used was verified experimentally by observing smoke in a setup in which the outer electrode and shield were replaced by clear plastic parts.

The ion current was measured with a model V, micro-microammeter of Beckman Instruments, Incorporated, Fullerton, California, which also incorporated a regulated 210-v potential source. Variable potential for the ion chamber was derived from this regulated voltage by means of a standard potential dividing circuit. The current reading of the micro-microammeter was simultaneously recorded.

Ions of each material tested were collected with a thermal precipitator, a Thermopositor of the Roy A. Martin Company, Atlanta, Georgia. Size distributions were determined from electron photomicrographs of the deposits.

Nuclei size measurements were made at ambient laboratory temperature, approximately 25C.

The experimental method was essentially that of Israël [4] and of Junge [2]. The method of data interpretation differed, however, in that individual ion sizes were examined, whereas previously, average data for an entire size distribution were obtained. The recorded information was plotted as ion current *versus* the voltage applied to the ion chamber at constant humidity giving curves all of which attained the same maximum ion current. Analysis of these

curves to give size distribution as a function of humidity was achieved by noting that the intercept on the ion-current axis of the tangent to the current-voltage curve divided by the current maximum gave the fraction of particles of the size completely collected at the voltage corresponding to the point of tangency. The relationship between collector voltage and size was calculated from the geometry of the ion chamber and Stokes' law relating mobility to ion size (assumed spherical). Because of the small ion size, corrections were applied for the effective viscosity of air. Complete details of the calculations are reported elsewhere [5].

Fig. 2, presenting curves for different size crystals of each material, gives the experimental results. As may be seen, non-hygroscopic particles attract relatively little water vapor up to the limits of the experiments. Hygroscopic nuclei, on the other hand, pick up (or lose, in the case of decreasing humidity) large quantities of water and change size in a characteristic manner. Moving from a lower to a higher humidity such nuclei undergo an abrupt size increase as the solid dissolves becoming a droplet. At still higher humidities the droplet picks up moisture in ever increasing

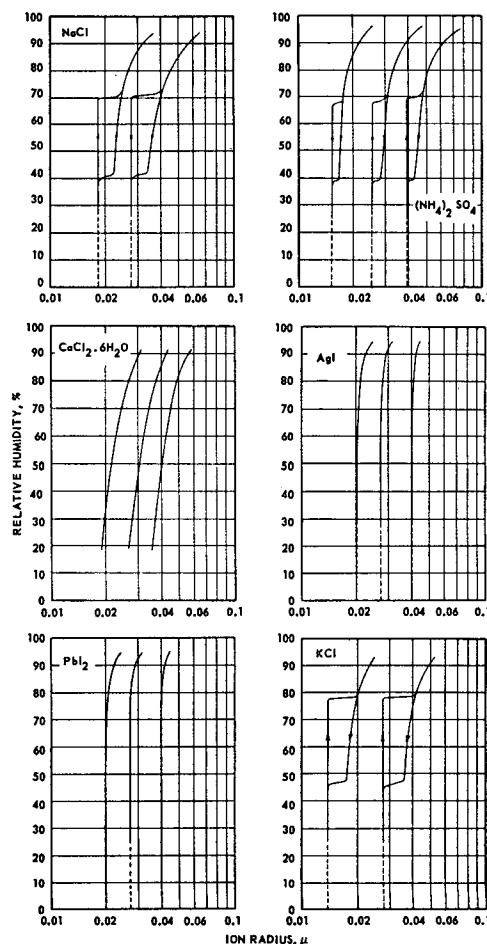


FIG. 2. Ion size as a function of relative humidity.

quantities. When humidity is reduced the droplet loses water just as before it gained. It does not, however, recrystallize at the humidity at which it dissolved. The final step back to the original crystal size takes place at a considerably lower humidity.

Solution and crystallization occur at conditions which are characteristic of the material, and both processes occur at lower and lower relative humidities as size decreases. For example, an $0.020\ \mu$ radius NaCl nucleus dissolves when the relative humidity reaches barely 70 per cent whereas a crystal of about $1\ \mu$ radius dissolves at about 74 per cent.

The pickup of moisture by droplets containing small amounts of dissolved material, *i.e.*, droplets at relatively high humidities, is in accordance with Köhler's theory as a comparison of the experimental curves given here with ones calculated as outlined by Howell [6], for example, will show. It should be noted, however, that many nuclei below about $1\ \mu$ in size have solubilities in excess of normal solubility and hence form droplets at relative humidities below that prevailing over saturated bulk solution. This means that at moderate humidities droplets composed

of supersaturated solutions are involved. For the most part, data on the properties of such solutions are unavailable and it was found necessary to obtain approximate values of the vapor pressure in these regions by calculation and extrapolation. Details are given elsewhere [5].

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