Electrofreezing of Supercooled Water[1])

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Summary – The results of a wind tunnel and cold chamber investigation on the effect of intense electric fields on ice-nucleation in supercooled water are presented. It is found that electrostatically charged surfaces and externally applied electric fields significantly enhance ice-nucleation. This finding is supported by a review of recent work reported in literature. Several mechanisms are considered which can be made responsible for electrofreezing. A discussion of the electrofreezing effect suggests that it is the electrical relief of the surface of a solid substrate rather than the crystallochemical relief which is the determining factor in heterogeneous ice nucleus formation.

1. Introduction

An excellent summary of work which was reported in literature on various topics in cloud physics has recently been given by MASON [1][3]). From this summary it is evident that ice formation in the atmosphere is a heterogeneous process initiated by ice forming nuclei whose effectiveness depends on the following factors: (1) on the size of the nucleus, i.e. the total surface area available for nucleation, (2) on the water solubility of the nucleus, i.e. the capability of the nucleus to exhibit a rigid, compact surface when in contact with water, (3) on the chemical nature of the nucleus, i.e. the type of atoms lying in the nucleus surface and the type of bonds which these atoms extend, (4) on the crystallographic nature of the nucleus, i.e. the symmetry of the geometric arrangement of the atoms and the distances between them, and (5) on the surface microstructure of the nucleus, i.e. the type of lattice dislocations, steps, cracks and impurity sites present at the nucleus surface. While all of these factors play some role in the initiation of ice from the vapor or supercooled liquid, none of these factors can uniquely be correlated to the effectiveness of an ice forming nucleus. It is the purpose of this paper to show that there is yet another and possibly most fundamental factor that controls ice-nucleation.

2. Experimental

a. *Wind tunnel studies*

As part of a detailed investigation on the effectiveness of clay particles as ice-forming nuclei, to be reported elsewhere, we studied the ice nucleability of substances

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[3]) Numbers in brackets refer to References, page 633.

which from literature were known to have poor ice-nucleating properties. Among those substances amorphous sulfur proved to be the most interesting one. For this study drops of highly purified water were freely suspended in the nuclei-free air stream of the UCLA cloud tunnel. This facility was described in detail by PRUPPACHER and NEIBURGER [2] and BEARD and PRUPPACHER [3] for stably suspending water drops in an airstream of temperatures warmer than 0°C. For stably suspending water drops and ice particles at temperatures colder than 0°C a specially constructed tunnel section was mounted inside the observation section of the main tunnel. This inner tunnel consisted of aluminum with two glass walls as does the main tunnel but had smaller dimensions such as to leave an air gap between them. As a result, the air stream temperature in the inner tunnel was uniform and the same as the walls of the inner tunnel. Since the relative humidity of the airstream varied between 60 and 80% the temperature of a suspended drop was somewhat lower than that of the airstream due to evaporation. However the effect of evaporation on drop temperature as a function of the Reynolds number of the drop is well known from the recent work of BEARD and PRUPPACHER [4] so that the actual drop temperature could easily be calculated. When a supercooled drop froze the transition period of phase change from water to ice was characterized by an abrupt lowering of its terminal velocity and often a tendency for the particle to drift horizontally. These motions were counteracted by lowering the airstream velocity and adjusting the inner tunnel which, by means of micrometer screws, could be tilted in any direction with respect to the main tunnel. Within a few seconds the all-ice particle would assume a characteristic mode of fall and in this way could stably be suspended in the airstream for any length of time. By means of a microsyringe drops of highly purified water with sizes between 100 and 350 μ radius were injected into the tunnel airstream and supercooled to a desired temperature while freely suspended. The purity of the water and airstream were such that none of the drops froze at temperatures warmer than -20°C. After a particular drop had come into equilibrium with the airstream, a small cloud of finely ground, dry amorphous sulfur was injected into the airstream well upstream from the drop's position by means of a specially constructed aerosol dispersing mechanism. A second or so later the sulfur particles, which had sizes with a mode at 1 to 2 μ diameter, passed through the observation section many of them colliding with the drop. Contrary to all expectations we observed that drops froze at temperatures as warm as -8°C by contact with the sulfur particles. On the other hand, drops formed from aqueous suspensions of sulfur particles froze at the same temperature at which pure water drops froze, indicating that sulfur particles embedded in a drop have poor ice forming capability. Indeed, one would expect that amorphous sulfur, whose crystalline nature is completely different from that of ice and whose chemical bonds have no similarity to the hydrogen bonds in ice, has poor ice nucleating properties. On the other hand, it is well known that dry sulfur particles become negative electrically charged when mechanically dispersed in air. One is therefore tempted to attribute the excellent ice-nucleability of sulfur particles on contact with supercooled water drops to an electric effect.

b. *Cold chamber studies*

Two experimental investigations were carried out in a large cold chamber. In the first study a highly polished steel sphere of 6 cm in diameter was covered with a thin polyethylene sheet in order to render the sphere's surface hydrophobic. The sphere was connected to the negative pole of a high voltage dc-power supply continuously variable between 0 and 30 kV. At a radial distance of about 2 cm from the sphere surface a half-hemispherical fine-mesh copper-wire net was mounted and connected to ground. The whole set-up was located inside a large walk-in cold chamber where the air temperature was set to about $-10\,°C$. Drops of highly purified water and of about 1 mm in diameter were placed onto the hydrophobic sphere surface. After the drops had come into temperature equilibrium with the environment the sphere was raised to an electric potential of -20 kV. A rise of the electric potential of the sphere was immediately followed by freezing of all drops sitting on it. In the absence of the electric field a population of the water drops froze at temperatures between -12 and $-24\,°C$ with a medium freezing temperature of $-18\,°C$. With this set-up the electrofreezing of drops was repeated many times with the same result. If the sphere was charged with -20 kV but the grounded electrode was removed, the drops did not freeze but froze immediately, from the drop base up, as soon as they were moved slightly by means of a small teflon rod. Mechanical movement of the drop in the absence of any electric field caused no freezing.

In the second study water drops of about 1 mm in diameter were placed on the surface of a solid polyethylene plate and cooled in the walk-in cold chamber to $-10\,°C$. The plate was then tilted and by means of a mechanical jerk the drops were suddenly caused to move and roll down the plate. By doing so they did not freeze and simply left behind a trail of unfrozen water beads. The set-up was then altered and the polyethylene plate carefully dried before the drops were placed on it. After the drops and plate were cooled to $-10\,°C$ the drop free portion of the surface was strongly rubbed by means of a specially cleaned and dried woollen cloth. This produced a strong electrostatic surface charge which was easily documented by placing the probe from an electrometer near the surface. If the plate was tilted all drops immediately froze while being forced to move and roll over the surface. Without an electric surface charge the drops would freeze at temperatures between -12 and $-24\,°C$ with a median freezing temperature at about $-18\,°C$. Indeed, from the pronounced chemical and crystallographic dissimilarity to ice one expects that polyethylene itself has poor ice forming properties. One is tempted therefore to attribute the excellent ice-nucleability of a highly charged polyethylene surface to an electric effect.

3. Review of experiments on electrofreezing reported in literature

It is the purpose of this section to show that our results are consistent with the results of similar experiments reported in literature. DUFOUR [5] was probably the first

to observe that freezing could be initiated in supercooled water by means of an electirc field. Unfortunately he gave only an insufficient description of his experiments so that no firm conclusions can be drawn. Many years later RAU [6] carried out the first quantitative electrofreezing experiment by studying the freezing of supercooled water drops in the presence of electric fields. The water drops had a diameter of a few millimeters and were placed a on highly polished chromium plate which served as a negative electrode. A metal pin or a sphere covered with glass or amber touching the top portion of the drop served as the second electrode. Between the two electrodes electric fields of 20 to 60 kV/cm were produced. By this set-up drops froze immediately at a temperature as warm as $-4\,^{\circ}C$ when they were exposed abruptly to electric fields of these intensities. SCHAEFER [7] froze supercooled water drops resting on plastic surfaces by exposing them to the spark from a Tesla coil. Similarly, SALT [8] initiated freezing in supercooled drops by means of an electric field between an aluminum foil, on which the drops were placed, and a wire probe about 1 cm above the drops. A detailed investigation on the electrofreezing effect was carried out by PRUPPACHER [9, 10]. While he verified that ice formation in drops which were supercooled by a few degrees freezing could indeed be initiated by electric discharges, he showed that the electrofreezing effect is not uniquely dependent on an electric discharge. He encased water drops of about 1 mm in diameter in hydrophobic polyethylene tubes and placed those between two plate electrodes which were submerged in oil. He showed that these drops froze at temperatures as warm as $-5\,^{\circ}C$ immediately after an electric field of 3 to 25 kV/cm was applied across the electrodes. No electric discharges took place during this experiment. On the other hand, a water drop deformed in the electric field and spread into a thin wedge at the triple interphase water-polyethylene-oil or air, while moving slightly across the hydrophobic surface. If instead of a small drop of water a column of water contained in a long polyethylene tube was placed between electrodes the water in the tube would not freeze down to a temperature as cold as $-18\,^{\circ}C$ no matter how intense the externally applied electric field was. Below $-18\,^{\circ}C$ the water in the tube froze as a result of nuclei on the tube wall and in the water. Most phases of the work of Pruppacher were independently checked and all the findings confirmed by EVANS[4]) in the Cloud Physics Laboratory of Professor B. J. MASON at Imperial College, London, England. ROULLEAU [11], ROULLEAU and POC [12] and POC [13] carried out their studies on the electrofreezing effect in a cloud chamber. They reported that large numbers of ice crystals formed in a supercooled cloud when a strong electric field of several kilovolts per centimeter was applied via two metal grids located in the cloud. GARRAUD [14] claimed that the results of Roulleau and Poc were due to formation of frost on the electrodes. Indeed, SCHAEFER [15] photographically presented evidence for the formation of a stream of ice crystals from the tip of very fine wire when it was electrically raised to a potential of 3 kV. In a more detailed experiment, ROULLEAU *et al.* [16] found that a clean wire electrically charged to a potential of 8 kV

[4]) L. F. Evans, Res. Rept. Cloud Phys. Dept., Imperial College, London, England (1962).

in the presence of a supercooled cloud would not produce ice crystals down to temperatures as low as $-20\,°C$. If, however the wire was covered with a hydrophobic substance a stream of ice was generated. These ice crystals resulted from supercooled cloud drops which were attracted by the wire in the asymmetric electric field and made contact with the hydrophobic surface where they froze and partially disintegrated into ice fragments. Ice nucleation on a hydrophobic wire was observed at temperatures as warm as $-5\,°C$. ABBAS and LATHAM [17] studied the electrofreezing of water drops of 1 to 2 mm in radius which were hung from a hydrophobic teflon rod. There the drops either were exposed to intense electric fields which caused the drops to disrupt, or they were pierced by electrically charged and uncharged nylon rods. Drop disruption as well as piercing was frequently followed by drop freezing when the drop temperature was below $-4\,°C$. SMITH *et al.* [18] extended Abbas and Latham's studies to drops which freely fell in air through electric fields whose intensity exceeded that necessary for drop disruption. Drop disruption was followed by drop freezing if the drop was supercooled to a temperature below $-6\,°C$. Similar results were found by KOENIG [19] who freely supported drops in the airstream of a wind tunnel and found that bag-break-up of supercooled drops was often followed by drop freezing. A completely different set of experiments was carried out by GABARASHVILI and GLIKI [20] who studied the freezing behavior of a volume of 1 cm^3 supercooled water into which crystals of naphthalene where dropped. The pure water with or without uncharged crystals always froze at a temperature of about $-5\,°C$ and began freezing from the vessel walls. In contrast to that, if the crystal was electrically charged freezing always took place as a temperature as warm as $-3\,°C$ and started immediately at the surface of the crystal a soon as it touched the supercooled water surface.

4. Significance of the electrofreezing effect to precipitation formation in atmospheric clouds

Before we shall attempt to suggest physical mechanisms which lead to electrofreezing we briefly want to consider whether electrofreezing as found in the laboratory is likely to occur in atmospheric clouds and whether it could significantly effect precipitation formation. The results of the laboratory experiments which we presented in the previous section showed that electrofreezing results when supercooled drops either become subjected to external electric fields of several kilovolts per centimeter, or collide with a dry hydrophobic electrically charged, solid surface. It is well known from field observations, that electric fields of several kilovolts per centimeter are only present in cumulonimbus clouds of mature thunderstorms. In such clouds also ice particles are usually found in large numbers so that it will be of little significance to the ice crystal budget of the cloud whether or not a few more ice particles are formed by electrofreezing. In such clouds also the growth of cloud particles to precipitation particles is well underway so that the field induced break-up of drops into frozen and unfrozen fragments hardly will affect the overall precipitation rate,

except that it may initiate local rain gushes which indeed have been observed to take place just following the build up and break-down of high electric fields. In the atmosphere solid surfaces are only present in form of aerosol particles. Except in air over urban areas the air of the free atmosphere does not consist of hydrophobic particles but rather of mixed particles with hydrophilic, water soluble compounds and partially wettable, water insoluble substances. In atmospheric clouds where the relative humidity is mostly between 97 and 102% all of these particles are in volved to various degrees in the condensation process and thus are located either inside cloud drops or ice crystals or are at least partially covered with a few layers of water molecules. However, near the edges of a cloud where downdrafts prevail dry particles may be mixed into the cloud air by turbulent eddies. Since there are a large number of ions inside and outside a thunderstorm cloud it is likely that some of these aerosol particles may be highly charged. Such particles could conceivably initiate electrofreezing. Charging of particles would be particularly high near lightning channels from where large numbers of ions and electrons diffuse out to attach themselves to aerosols. The laboratory results presented in the previous section suggest also that cloud seeding would be much more efficient in the presence of electric fields, or if the particles used for seeding are electrically charged.

5. Possible mechanisms which underly the electrofreezing effect

It lies near to assume that electrofreezing as observed in the laboratory is merely an art-effect caused by fresh nuclei which are formed from electrode material during electric discharges between the electrodes. Indeed ALBANI [21] found that large numbers of ice forming nuclei were produced during spark formation between silver electrodes. Using this point of view BLANCHARD [22] criticized the conclusions of SALT and others who assumed that electrofreezing is a result of dielectric polarization. There is no doubt that under certain conditions electrofreezing may be caused by artificial production of nuclei resulting from electric discharges. However, the present experiments presented in Section 2 of this article and those of PRUPPACHER [9] showed clearly that the electrofreezing effect is not dependent on electric discharges.

A second mechanism was suggested by LOEB [23] based on PRUPPACHER's [9] observation that freezing of supercooled water drops was intimately related to the presence of an external electric field of sufficient intensity to deform the drop and to draw portions of it out into a thin wedge or spindle. Based on his earlier work [24] LOEB conjectured that when a drop becomes sufficiently deformed such that it is drawn out into a thin, liquid filament of such small dimension that the surface tension forces are no longer able to govern the drop shape (below a diameter of about 10^{-6} cm) the liquid aggregates take the form of minute crystallites which might act as excellent ice-forming nuclei. Thus, Loeb argued that for electrofreezing to take place (1) the drop has to be drawn by the electric field into a fine point which conceivably has crystallite size, (2) a heat conducting surface has to be situated adjacent to the liquid filament

so that the latent heat liberated during freezing can readily be dissipated, and (3) sufficient free space has to be available in the vicinity of the liquid filament to accommodate the expansion of the water while freezing. Even though all three requirements were fulfilled in Pruppacher's experiments we shall give two arguments to show that Loeb's mechanism is highly unlikely to explain electrofreezing. (1) X-ray diffraction studies reported in literature show that the radial distance to neighboring water molecules from a given water molecule slowly approach those in ice as the temperature decreases below 0 °C. Numerous scientists deduced from this finding that the structure of liquid water has ice-likeness and becomes increasingly ice-like at lower temperatures. However, we need to realize that the results derived from X-rays only apply to the mean state in water and does not predict how the molecules are grouped at any one time. An attempt to describe the structural evolution in time of water molecules in liquid water between +35 and −10 °C has very recently been made by Rahman and STILLINGER [25] who simulated the liquid water by an electronic computer using the molecular dynamics method. This method allows studying a system of a few hundred water molecules as it evolves structurally in time by the laws of classical dynamics subject to an effective pair potential that incorporates principal structural effects of many-body interactions in real water. This study showed that there is a high degree of disorder in the 3 dimensional network of water molecules with mostly random molecular configurations which are rather homogeneous in density. While the bonds between neighboring water molecules tended to be tetrahedrally oriented the average degree of bending away from bond linearity and ideal approach directions was considerable. Although distorted polygons of 4, 5, 6 and 7 molecules appeared, no recognizable clusters of ice-like type or clathrate-type, or networks with interstitial molecules were found. Even at a temperature −8 °C this situation did not change significantly in that nowhere organized beginnings of ice like clusters were found, except that the bonds between neighboring molecules seemed to be more linear and the bond angles closer to tetrahedral angles than at warmer temperatures. The most significant finding was the presence of numerous dangling OH bonds which persisted for times longer than the vibrational period of water molecules. Thus, it appears from this most recent study that small water quantities do not contain ice like clusters or crystallites which could act as ice nuclei, as was envisioned by Loeb. (2) Even if ice-crystallites would exist in water they would have to have a radius larger than 10^{-6} cm before they could serve as an ice-germ in equilibrium with a water environment of a temperature between −4 and −5 °C at which electrofreezing was initiated by PRUPPACHER [9]. This is evident from the result of DUFOUR and DEFAY [26] who determined that the equilibrium temperatures for ice-germs of 10^{-6} cm, 5×10^{-7} cm and 10^{-7} cm radius are −4.2; −8.3, and −37.9 °C respectively. The two arguments presented above suggest strongly that electrofreezing cannot be explained on the bases of the mechanism suggested by Loeb.

A third mechanism for electrofreezing was suggested by SMITH *et al.* [18]. These authors proposed on the bases of their experimental results that during an abrupt

disintegration of a water drop in an external electrical field a cavitation bubble is produced in the water which on collapsing induces freezing. While from this and other evidence presented in literature it cannot be contested that cavitation in supercooled water and other supercooled melts does, under certain conditions, initiate solidification, it must be pointed out that during the present electrofreezing experiments and during those of Pruppacher, Rau, and Roulleau and coworkers, presented in Section 3 of this paper, the deformation of the water drops was small and not followed by disruption. Cavitation can therefore not be made responsible for their observations.

A fourth mechanism for electrofreezing was suggested by Rau, Salt and by Gabarashvili and Gliki. These scientists proposed that electrofreezing of supercooled water is due to dielectric polarization of water in high electric fields. This idea was discounted by PRUPPACHER [9] who showed that freezing could not be initiated im columns of supercooled water by means of an external electric field even if the intensity of the electric field was as large as 30 kV/cm across the water column. Indeed, simple calculations show that electric fields of the order of 10^6 V/cm rather than 10^4 V/cm are needed in order to significantly orient watermolecules in an electric field. However, Gabarashvili and Gliki pointed out that the electric field could reach such large values over very short distances at the surface of highly charged particles which have emerging, pointed surface features. At such projections water molecules would become strongly bound to the surface, at least in the first layer adjacent to the solid surface. This in turn would stabilize the water structure farther out and this be conducive to freezing. Form a microscopic determination of the dimensions which characterize the projections on the naphthalene particles which were used for the electro freezing experiment, Gabarashvili and Gliki concluded that the electric field near the projections was as high as 7.2×10^6 V/cm on naphtalene particles which were charged to a potential of 3000 V. Based on knowledge available in literature on the structure of water and based on classical nucleation theory they computed that the ice-nucleation rate was approximately 250 times enhanced on electrically charged than uncharged particles. The argument of Gabarashvili and Gliki is, at least in some respects, objectionable in that ice formation is not only determined by energy criteria but also by steric criteria. It is for instance well known that ice formation does not necessarily start in very strongly adsorbed layers of water molecules where an ordered state completely different from that of ice prevails.

It is in this context that a freezing mechanism suggested by EVANS [27] is of importance. From his extensive nucleation studies Evans came to the conclusion that in supercooled water 3 dimensional (3D-) ice nucleation does rarely take place directly on the surface of a solide substrate but rather on top of water layers already adsorbed on that solid substrate. Below a certain temperature such an adsorbed layer may normally transform from an initially disordered state into an ordered state (2D-ice) upon which 3D-ice will readily nucleate. Many solid substrates have the tendency to adsorb under equilibrium conditions water molecules in a firm oriented layer with a structure very different from that in bulk water or bulk ice. This situation, for instance prevails

at plastic surfaces of polymers with non-polar molecules around which water adopts a strongly bonded structure which often results in clathrate formation with increased hydrogen bonding but decreased ice-likeness [28, 29], or at surfaces of silicates where the adsorption of the first layer of water is not governed by the similarity between the silicate and the ice crystal lattice but by the randomly distributed, strongly hydrophylic, and therefore strongly adsorbing, exchangeable cations on the cleavage faces. Based on his own experiments Evans proposed that under such conditions a relatively large supercooling is needed to transform an adsorbed layer into 2D-ice. If such surfaces are submerged in water they exhibit relatively poor ice nucleating properties. This notion is also in agreement with the result of PRUPPACHER [9] who showed that the median freezing temperature of millimeter size drops on plastic surfaces, devoid of electric surface charges was about $-18\,^{\circ}C$, and the results of GOKHALE [30], HOFFER [31], and PRUPPACHER [unpublished] who showed that the median freezing temperature of drops made from clay suspensions was between -12 and $-18\,^{\circ}C$ if the drops had millimeter size, and between $-24\,^{\circ}C$ and -32 if the drops had radii of about 110μ. However, if water freshly wets a dry surface of a water insoluble substance, as is the case during the initial moments of contact between such a surface and a supercooled water drop the adsorbed layer stays disordered for a short period of time. Evans argued that transformation of such a layer into 2D-ice is energetically easier and therefore takes place at a much smaller supercooling than transformation of an adsorbed layer in equilibrium with bulk water. Under such conditions a surface may exhibit relatively good ice-forming characteristics. In order to support his proposed mechanism, Evans cited the experiments of PRUPPACHER [9] who froze drops on plastic surfaces at temperatures as warm as $-5\,^{\circ}C$ when these were suddenly deformed and forced to move slightly be means of an external electric field. Further support of Evans' mechanism comes through the experiments of GOKHALE and SPENGLER [32] and GOKHALE and GOOLD [33] who found that water drops froze at temperatures between -3 and $-8\,^{\circ}C$ just after they made contact with dry particles of clay and other silicates. Also our present experiments with sulfur particles and plastic surfaces support the Evans mechanism.

Thus, the available experimental evidence suggests that even under transient conditions the probability for nucleation of 2D-ice at a solid surface in contact with supercooled liquid water is considerably larger in a disordered adsorbed layer than in a firmly bonded adsorbed layer. This is reasonable if we consider that a disordered adsorbed layer may be characterized by a network of water molecules where some portions are anchored by a few of the best adsorbing sites, where, a tendency to tetrahedral coordination still exists, and where structural fluctuations take place due to thermal agitation. In a firmly bonded adsorbed layer, on the other hand, all the molecules are locked into fixed positions thus preventing structural fluctuations to nucleate 2D-ice.

It is interesting to note that silveriodide also behaves in such a manner despite its crystallographic structure which is closely related to that of ice. Hoffer found

that the median freezing temperature of drops of 110μ radius made from aqueous silveriodide suspensions was −15°C while Gokhale and Spengler showed that dry silveriodide particles froze supercooled water drops on contact at temperatures between −2.5 and −4°C.

If we assume that the Evans mechanism indeed describes the basic features of heterogeneous freezing of water it is but a small step to extend this mechanism to include electrofreezing. It is well known that any dry, dielectric surface becomes electrically charged and develops an intense electric surface field as a result of mechanical rubbing. Thus, by asymmetric rubbing on each other, dry dielectric particles become electrically charged during their dispersal in air [34], the electric surface field being in particular intense near emerging, pointed surface irregularities. Very intense, local electric fields develop also in the presence of an external electric field at the tip of spindle or wedge-like deformed drops of water which under this condition has to be considered as an electric conductor. During the transient wetting of a surface an intense local electric field serves as highly active site. Such a site, in turn, may act on the dangling OH-bonds, which are present in water according to Rahman and Stillinger, by linking some of them through bond polarization to other water molecules thus increasing the total number of hydrogen bonded water molecules, and by linking some others to the solid surface thus anchoring the water network. It is reasonable to expect that under such conditions nucleation of 2D-ice and thus freezing is promoted. This is in agreement with our present experimental results and with those of PRUPPACHER [9].

Up to now we have discussed the effect of electric fields on ice formation from supercooled water. In this context it is useful to ask whether electric fields also effect the nucleation of ice from the vapor. Unfortunately, no studies have been carried out on this problem. However, CHOPRA [34, 35] and MIHAMA and TANAKA [36] studied the deposition of gold and silver from the vapor on a glass or NaCl substrate. Their studies showed that the new phase was deposited in the form of islands which grew by lateral coalescence with other islands producing large islands considerably faster if an external electric field was applied across the substrate then if there was no field. It is well known from numerous studies that ice is also deposited from the vapor in the form of islands on a substrate. Thus, by analogy we suspect that electric fields will increase the size of the water molecule clusters and in this way enhance the ice nucleability of that substrate.

Having demonstrated the profound effects of electric fields on ice nucleation we need to reconsider the completeness of our list of characteristics for a good ice-forming nucleus. From literature evidence it seems unquestionable that each of these crystallochemical characteristics indeed do play some role in the ice nucleation mechanism. However, in the light of our discussion presented above we suspect that the crystallochemical characteristics are of considerably smaller importance than the electrical characteristics. In a series of very recent articles (which due to lack of space cannot all be listed here) DISTLER and co-workers [38], studying the deposition of gold and silver on various substrates, came to the same conclusion and pointed out that it si the elec-

trical 'relief' of the surface of a solid substrate rather than the crystallochemical which is the determining factor in heterogeneous nucleus formation.

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