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# **Freezing Water with Sized AGI Particles**

**a survey**

**Part 1**

**Atmospheric Science Laboratory**

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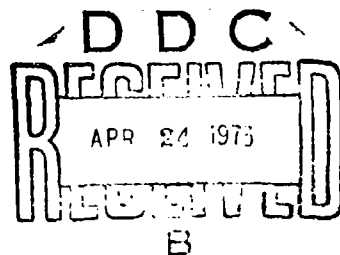
# **FREEZING WATER WITH SIZED AgI PARTICLES**

## **PART I: A SURVEY**

By

**Hermann E. Gerber**

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13. ABSTRACT A survey is made of previous experimental and theoretical attempts to determine the AgI particle size-ice nucleating activity relationship. This relationship is important since the successful phase transformation of water to ice in a supercooled cloud must depend on both the concentration and size of the injected AgI particles.  Fletcher's classical sublimation theory is found to be inconsistent with the few existing experimental results. The surprising conclusion is reached that a proper explanation of the behavior of AgI as a function of particle size has not been given. The Goetz Aerosol Spectrometer is suggested as the instrument capable of measuring the long elusive AgI particle size-activity relationship.		

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## INTRODUCTION

It is generally accepted that AgI in a finely subdivided form and suspended in air provides the most practical and efficient means of modifying supercooled clouds. Thus it comes as somewhat of a surprise that in more than 25 years since Vonnegut [1], [2] discovered the ice nucleating ability of AgI, no one yet has given a proven explanation of the behavior of AgI as a function of its subdivision. That information is of obvious practical importance. For most modifications the vastness of the atmosphere dictates that the output of AgI particle generators, which can vary over as many as four orders of magnitude [3], be optimized. In addition, others [4], [5] have suggested that a numerical value of the efficiency is also required, since the outcome of the cloud seeding depends on the in-cloud concentration of the AgI particles.

At first glance the largest number of AgI particles capable of nucleating ice simply might be synonymous with the greatest attainable state of subdivision of the bulk AgI in the generator. Unfortunately, that is not the case. There must exist a particle size below which the particles' curvature and limited surface area inhibit nucleation. A further complication is the need to specify which of several nucleating mechanisms are involved in the cloud, and how those are dependent on the particles' size. Theoretical and experimental attempts have been made to determine the AgI particle size-ice-nucleating activity relationship. However the theory has not yet been proved correct, and the experiments have been very limited. This paper, the first of a four part series, introduces our effort towards filling this gap in our knowledge.

## FLETCHER'S THEORY

For a time it seemed that Fletcher [6], [7] had developed a successful theory. He essentially applied the classical nucleation theory of Volmer and Weber [8] to AgI particles which had a uniform crystalline surface and which nucleated ice by sublimation.\* The theory also required specifying a value for the

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\*The term sublimation, as used by Fletcher, strictly refers to the process in which ice nucleates on the particles directly from the surrounding vapor. In this paper sublimation will also include ice nucleation at and below water saturation without the presence of macroscopic quantities of water. The adsorption of water onto the AgI surface to form liquid embryos which subsequently freeze is thus included. Freezing will refer to ice nucleation when the AgI particle either contacts or is enclosed in a water droplet.

unknown contact angle of ice on the AgI surface. Fletcher assumed that all particles larger than  $2\mu\text{m}$  in diameter nucleated ice by sublimation at the observed activity threshold of about  $-40$ . He then used his theory to calculate an effective contact angle. The completed theory predicted that a different threshold temperature existed in the nucleation ability for each size of AgI particle. Fletcher used those thresholds to compute his familiar activity curve where the  $\log_{10}$  of the number of active particles per gram of AgI is given as a function of subfreezing temperature. The theory found strong support from the activities of AgI smokes measured in laboratory cold boxes. Often the experimental curves roughly paralleled the theoretical curve, while at most the difference between the two was several orders of magnitude (e.g., Vonnegut [2], Grant and Steele [3], Smith and Hoffernan [9], and Fluckay and Wells [10]). This apparent agreement suggested that the assumptions of the theory were essentially correct. Fletcher's contention that the AgI acted as a sublimation nucleus was also supported by the earlier experimental observations of Twilong [11], Schaefer [12], and Birstein [13] which showed that AgI demonstrated activity below water saturation. In addition, Bigg [14] and Hoffer [15] respectively found the AgI nucleation by freezing threshold at  $-120$  and  $-210$ , thus confirming for a time that sublimation was the predominant nucleation mechanism.

Subsequently a large amount of experimental work suggested that Fletcher's theory had serious flaws. The action of AgI as a sublimation nucleus was initially challenged by other measurements of the freezing threshold. Numerous workers (e.g., Boyardelle [16], Rouilleau [17], and Mason and Van der Heuvel [18]) found a freezing threshold of about  $-40$ . Edwards and Evans [19] attributed Bigg's and Hoffer's colder values to the depression of the freezing threshold by the presence of iodide ions in their supercooled water. Early work [18] gave the sublimation threshold as  $-120$ . However, it has since been reconfirmed [20], [21] that this threshold also is about  $-40$ .

The sublimation mechanism was questioned further by the increasing evidence that AgI acts primarily as a freezing nucleus. Edwards and Evans [22] experimentally demonstrated how small AgI particles were efficient nucleators only if they were enclosed or rare in contact with supercooled water. They also found that about 110% relative humidity was required to induce nucleation if the particles did not see water. Since the relative humidity in natural clouds rarely exceeds 101%, they concluded that the nucleation of ice must depend primarily on the collision of the

Agl particles with the supercooled cloud droplets, which then froze. The work of many [20], [23-28] supported that conclusion. Still, much more quantitative data is required to clarify the role that each nucleation mechanism plays as a function of incloud environment. Nevertheless, the evidence of the -4C freezing threshold and the observed importance of the freezing mechanism suggest the possibility that the many experimental activity curves which have been drawn next to Fletcher's sublimation theory curve really reflect the freezing and not the sublimation behavior of AgI.

The concept of threshold temperature in Fletcher's theory was shown to be invalid [23], [24], [29]. According to his theory, the nucleation rate of particles of a given size is either negligible or so large that all particles activate. This rapid change in the rates occurs at an amount of supercooling termed the threshold temperature. Edwards et al. [23] observed the ice nucleation of monodisperse AgI particles in water, but did not find threshold temperatures for the three particle sizes, 0.17, 0.75, and 3.5 $\mu$ m, studied. Instead they determined that a fraction of the particles of each size activated and that the activity was proportional to the particles' surface area for any given temperature. Katz [29] and Mossop and Yayaweera [24] observed the same effects with sized AgI particles in a cloud chamber.

It had not been pointed out in the literature that the concept of threshold temperature is also inconsistent with the nucleation time lag observed first by Vonnegut [2] and again by Warburton and Jefferran [30]. AgI particles were found to nucleate ice in the cold chamber in decreasing amounts, even after 50 minutes had elapsed. Since the theoretical nucleation rate for a given size of AgI particles on either side of the threshold temperature is either immense or negligible, those particles with smaller sizes should not activate at all, while those with larger sizes should appear almost instantaneously. Thus a theoretical time lag is not possible. Fletcher's [7] explanation of the time lag was inconsistent with his own theory. He felt that the different nucleation rates of polydisperse AgI particles accounted for the experimental observations.

The temperature dependence of the time lag suggested to Edwards and Evans [20] that a possible explanation was found in the longer time required for the better and more hydrophobic nucleation sites to become wetted at the warmer temperatures. Vonnegut [2] explained the time lag as due to the chance occurrence of creating a stable ice embryo on the AgI surface. He felt that the temperature

dependence of the time effect ruled out, as an explanation of the time lag, the additional time required for the particles to diffuse to the droplets. It is interesting to note how the majority of experimental work with AgI has ignored the early observations of nucleation time dependency.

The failure of Fletcher's theory to account for the observations of fractional activity and the dependence of activity on particle area was blamed on his assumption that any part of the uniform surface of the AgI particle was as likely as any other part to nucleate the ice phase. Edwards et al. [23] neatly accounted for the observations by postulating instead the existence of an inhomogeneous AgI surface on which randomly distributed sites of widely different efficiencies nucleated ice. Various proposals have been made about the unknown nature of these "active sites," including hygroscopic patches of impurity atoms [31], [32], electrical inhomogeneities [20], and crystal growth steps [33]. Since the first two types of sites affect the adsorption of water on the AgI surface, they might well be important in nucleation by sublimation. However, their roles in freezing are uncertain. The third type of site was used by Fletcher [33] to develop new rate expressions for nucleation by freezing. He was now convinced that his earlier application of classical theory was incorrect.

Nearly all of the experiments referred to above have suggested errors in various aspects of Fletcher's original theory. Yet only a handful of work has been done on the most important one: the theory essentially tries to predict the size of the AgI particles below which they no longer assist nucleation. Probably that size as well as the size of most of the particles from AgI generators is below  $0.1\mu\text{m}$ . Thus measurements of the relationship between nucleation ability and particle size in that range are of the greatest relevance not only for comparison with the theory, but also for discovering the most active generator output.

Owing to the difficulty of determining particle size spectra below  $0.1\mu\text{m}$ , those measurements have not been completed. Some results were obtained by Maruyama [34], Mossop and Yayaweera [24], Sano et al. [35], and Edwards and Evans [22]. The first three investigators measured the nucleus size of replicated ice crystals formed by AgI smoke particles injected into a cold chamber. Maruyama found 0.4, 0.2, and  $0.08\mu\text{m}$  to be the sizes of the smallest active nuclei at -5, -10, and -14C, respectively, although the increments in his size histogram were about  $0.1\mu\text{m}$ . Mossop and Yayaweera tested AgI-NaI smoke and also found a temperature dependence, but shifted to smaller particle sizes. The involved nature of the replication technique appears to have made impractical

a thorough study of the activity-particle size relationship. The last two groups of investigators sized the AgI particles before testing their activity. Sano et al. found approximate agreement with Fletcher's sublimation theory, while Edwards and Evans concluded that particles of about  $0.02\mu\text{m}$  activated ice near  $-90^\circ\text{C}$  by the freezing mechanism. Two difficulties are encountered in sizing AgI particles beforehand. Not only is the definite connection between the particles' size and activation lost, but in addition a guess must be made of the nucleation mechanism when the particles are blown into the cold box.

#### CENTRIFUGAL SIZING OF NUCLEI

Allee et al. [36] proposed a new technique that promised to eliminate the experimental difficulties of sizing active ice nuclei. Essentially, the Goetz Aerosol Spectrometer, a high-speed centrifuge developed by Coetz et al. [37], is used to deposit airborne nuclei along its inert collection foil according to their size. The foil is removed from the centrifuge, cooled to the desired subfreezing temperature, and then humidified in a way which simulates the conditions for nucleation by either sublimation or freezing. In this controlled environment, the active nuclei grow into visible ice crystals. To find their size, only the location of the ice crystals need be recorded since the coordinates of the foil are related to particle size by Stokes law after calibration of the foil with particles of known size, shape, and density.

The centrifuge technique has been tried several times with AgI smoke particles. On two occasions (Allee et al. [38], and Gerber [39]) the measured activity spectrum of the AgI particles between the sizes of  $0.02$  and  $0.1\mu\text{m}$  closely resembled the predictions of Fletcher's sublimation theory. However, Gerber [40] later suggested that the freezing mechanism had actually been observed due to flaws in the experimental procedure. Additional measurements [41] of the particles' freezing activity gave results which differed somewhat from the previous work. Some uncertainty existed in these last measurements since the size of the AgI particles was difficult to define due to their highly coagulated nature. In addition, a large error might have been included since the nucleation time lag again was not accounted for. Obviously, the centrifuge technique had not yet been successfully utilized.

The Goetz Aerosol Spectrometer was chosen for the present study because it is the only available instrument capable of determining the small sizes of active AgI particles. Although the manufacturer of this centrifuge claimed sizing accuracy for polydisperse

particles between 0.03 and 3.0 $\mu$ m, controversy surrounded the accuracy of its measurements for many years. In a comprehensive theoretical discussion of the instrument's behavior, Stober and Zessack [42] showed that the centrifuge had inherent flaws which prevented it from accurately sizing particles. Although they offered a limited amount of supporting experimental work, some question of the instrument's capabilities still remained. They had not explained why the original calibration work of Goetz et al. [37] reflected proper operation of the centrifuge. Additional experimental evaluations of the centrifuge failed to clarify its usefulness. Raabe [43] sided with the theory, but the statistical error of his data was too large to permit a firm conclusion; the work of Baust [44] found abnormalities in the operation of the centrifuge, but not in the manner predicted by Stober and Zessack [42].

Gerber's recent evaluation of all the previous work concluded that the verdicts of the instrument's capabilities lacked experimental evidence [45]. As a result of the first thorough calibration, he discovered the operating conditions for accurate use of the centrifuge, the incorrect assumption which invalidated the theory of Stober and Zessack, and an explanation of the inconsistencies in the previous calibration attempts. The one other commercially available centrifuge [46] was not suitable for the present purpose, since it loses its resolution for particles smaller than about 0.1 $\mu$ m.

#### CONCLUDING REMARKS

The subsequent papers of this study will describe the use of the centrifuge technique to measure the ability of AgI smoke particles to nucleate supercooled water as a function of particle size, temperature, and activation time. The experimental aspects, generation of the AgI particles, measurement of their shape and size distributions, construction of a new cold chamber with the proper temperature and humidity control, and the measurement of the active fraction of the AgI particles will be given in detail. In addition, another look will be taken at Fletcher's theories to see how they fare in the particle size region where they should be most applicable.

It is hoped that this effort will answer some of the many remaining questions concerning the action of AgI as a freezing nucleus. Those questions include: What is the physical nature of the nucleation process, and can this process be described theoretically, what is the proper role of the nucleation time lag, and perhaps most important of all, what is the relationship between AgI particle size and nucleating ability?

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