

"REVIEWED"

GENERATION OF ICE NUCLEUS AEROSOLS BY SOLUTION AND PYROTECHNIC COMBUSTION

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Abstract. Combustion of acetone solutions of AgI and various chemical solubilizers and additives is used to generate ice nucleus aerosols for weather modification research, development and field use. Nucleus chemical compositions have varied from AgI itself to 2AgI-NaI, and 2AgI-KI, to Ag(Cl)I and to Ag(Cl)I-x NaCl (where x has varied from 0.125 to 5), over time.

Combustion of pyrotechnics to produce nucleus aerosols containing AgI is also widely used. Silver iodide and silver chloriodide "contact" nuclei have been the most often employed. More recent developments have led to the generation of Ag(Cl)I nucleants containing soluble salts, which function by rapid condensation-freezing mechanisms, rather than the cloud droplet "contact" mechanism.

These research and development efforts over a 50 years period have led to a better understanding of the relationships between ice nucleus compositions, activities, and the rates and mechanisms of ice crystal formation. Ice nucleus aerosols of optimum utility for field applications are now possible of generation.

1. INTRODUCTION

The chronology of the development of silver iodide containing chemical compositions and aerosols for use in weather modification activities traces an interesting sequence of mixed research and development efforts conducted over a period of fifty years. That a seemingly simple R and D effort should have taken this length of time is perhaps understandable if one considers the multi- and interdisciplinary nature of cloud seeding programs, the development of cloud chambers and alternate attempts to characterize these aerosols and the unfolding understanding of how various chemical variants of the basic AgI aerosol function in cloud. It would appear now, that the end of the program is in sight.

This article considers only pyrotechnic and solution combustion compositions and ice nucleus activity spectra that have been published and that the author is personally familiar with. Many other aerosol generation systems and devices are in current usage.

2. CHRONOLOGY

The discovery of the utility of silver iodide as an ice nucleus by Dr. Bernard Vonnegut at the General Electric Co., Schenectady, New York, (Vonnegut, 1848), followed the discovery by Vincent J. Schaefer that solid carbon dioxide ("dry ice") would induce the formation of ice crystals in a supercooled cloud (Schaefer, 1946). The initial aerosol generation system was the combustion of an acetone solution of silver iodide, using ammonium iodide (NH₄I) as the solubilizing agent (Vonnegut, 1949). The product of combustion was relatively pure AgI, contaminated only by trace amounts of hygroscopic salts from the original AgI preparation, which are co-precipitated with the AgI.

In 1950, however, Dr. Vonnegut published on an acetone combustion system in which sodium iodide, NaI, was used as the solubilizing agent for AgI in acetone (Vonnegut, 1950). The product aerosol was now 2AgI-NaI, rather than AgI, since NaI is not destroyed by combustion, as is NH₄I. The 2AgI-NaI aerosol, as cloud chamber studies at water

saturation in a droplet cloud have shown, was not an improvement over the AgI aerosol in terms of ice nucleus activity. These results however, do not imply that the 2AgI-NaI aerosol is less effective in atmospheric application than the AgI aerosol. Other than the occasional use of the 2AgI-KI aerosol from the AgI-KI-acetone combustion system, there were no changes in nucleus compositions from solution combustion until 1979.

The development of large cloud chambers, primarily at the Colorado State University, Department of Atmospheric Science, initiated the determination of ice nucleus activity spectra as a function of temperature in the 1960s, (Grant and Steele, 1966). In subsequent response to requests for ice nuclei with higher activities, the generation of AgI aerosols from the AgI-NH₄I-acetone-water combustion system was reintroduced in 1970 (Finnegan, et al 1971), and became the nucleus of choice for field programs.

3. SOLUTION COMBUSTION GENERATION

A discovery by Thomas W. Slusher, Nuclei Engineering Inc., Lewisville, Colorado, (Slusher, 1978) showed that the addition of an organic chlorine compound to pyrotechnic generators of AgI aerosols improved the activity of the aerosol nuclei significantly. This led to new research on ice nuclei generation by acetone solution combustion. In 1979 a study was initiated on the synthesis and characterization of AgI-AgCl solid solution nucleus aerosols by solution combustion. An increase in ice nucleus activity was expected for this nucleus composition, based on the work of Slusher, and on the prevailing concept of an improved structure match of nuclei with ice (Vonnegut 1947, Vonnegut and Chessin, 1977) that would result from the addition of chlorine atoms into the AgI structure lattice. The research program was designed primarily, however, to address the mechanism of ice crystal formation for this mixed AgI-AgCl nucleus, as well as AgI itself. (DeMott, 1982, DeMott et al 1983). The mechanism of ice crystal formation for these two nucleus compositions was unequivocally established as a sequence of steps starting with the Brownian scavenging of nuclei by cloud droplets, followed by droplet freezing, and the growth of ice crystals, at temperatures of from -5°C to -16°C.

The second phase in the study of the rates and mechanisms of ice crystal formation, and activities of ice nucleus aerosols at the cloud

simulation laboratory at Colorado State University concerned the 2AgI-NaI nucleus (Vonnegut, 1950). This nucleus aerosol, generated by combustion of its acetone solution, has been used world-wide since its disclosure and is still in use in several large field programs. Cloud chamber investigations (Blumenstein et al, 1987) demonstrated that, at water saturation in a droplet cloud, the ice nucleus activity of this nucleus was low at temperatures above -12°C, compared to the basic AgI aerosol (DeMott et al, 1983), its threshold temperature for activity was below -6°C and the rate of ice crystal formation for this nucleus was very slow. Cloud chamber tests showed that complete ice crystal formation and fallout required an hour or more at -10°C and -12°C. Correcting for aerosol dilution by cloud air introduction, the process should have taken about 2 hours. The mechanism of ice crystal formation was condensation followed by freezing as expected, but the reason for the very slow rate process and low activity was not elucidated until a later date. The second step in the formation of ice crystals, following water vapor absorption, is the actual ice nucleation step, slowed markedly by the large amount of NaI present. The large I⁻ ion concentration enhances water molecule dipole-dipole interactions, while suppressing the hydrogen bonded structure of water molecules associated with Ag⁺ ions on the AgI particle surface. This hinders and slows the nucleation mechanism (Finnegan 1998).

The third phase in the program was undertaken to overcome the low activity, poor threshold temperature and slow ice crystal formation rate of the 2AgI-NaI nucleus and produce a fast functioning, condensation-freezing nucleant with a high ice nucleus activity at temperatures above -12°C. Since the alkali metal iodides solubilize AgI in water at high concentrations (as well as in acetone) the difficulties were presumed to be due, at the time, to the alkali iodide present in relatively high concentrations in the 2AgI-NaI and 2AgI-KI nuclei, either destroying active sites or forming relatively stable complexes with the AgI (Burkardt et al 1970).

The alkali metal chlorides do not solubilize or form complexes with AgI, and methods of synthesizing composite nuclei compositions such as Ag(Cl)I x NaCl where x was varied from 0.5 to 5 were investigated (Feng and Finnegan, 1989). The NaCl was generated by adding sodium perchlorate (NaClO₄) in varying amounts to the AgI-0.5 NH₄I-NH₄ClO₄-acetone-water combustion solution of

DeMott (1983). During combustion, the NaClO_4 additive is reduced to NaCl . The mechanism of ice crystal formation was, as expected, found to be condensation followed by freezing. The optimal composition for activity and rate of ice crystal formation was found to be $\text{Ag}(\text{Cl})\text{I}-4.0 \text{ NaCl}$. Nuclei particles of this composition have salt quantities attached which can rapidly form dilute salt solutions of 10^{-3} molar concentration or less at water saturation. Particles with smaller quantities of attached salt form solution droplets of higher salt concentration, heading to lower rates of nucleation and lower yields of ice crystals. This is a manifestation of the Kelvin effect (Pruppacher and Klett, 1978). Composite nuclei of AgI or $\text{Ag}(\text{Cl})\text{I}$, and a soluble salt, have slower rates of nucleation than AgI or $\text{Ag}(\text{Cl})\text{I}$ nuclei (Finnegan 1998). The dramatic increases in activities and rates of ice crystal formation observed for this family of ice nucleus aerosols confirmed the hypothesis that $\text{Ag}(\text{Cl})\text{I}-x\text{NaCl}$ aerosols would be superior to $2\text{AgI}-\text{NaI}$ aerosols for field application, in terms of activities and rates of ice crystal formation. More recent studies have shown that for generation of $\text{Ag}(\text{Cl})\text{I}-0.5 \text{ NaCl}$ aerosol, combustion of acetone solutions of $\text{AgI}-0.5\text{NaI}-0.4 \text{ C}_6\text{H}_4\text{Cl}_2$ (paradichlorobenzene) is a better generation system. The $\text{AgI}-0.5\text{NaI}-0.4 \text{ C}_6\text{H}_4\text{Cl}_2$ acetone generator solution and the $\text{AgI}-0.5 \text{ NH}_4\text{I}-0.3 \text{ NH}_4\text{ClO}_4-0.5 \text{ NaClO}_4$ -acetone-water generator solution, on combustion, give identical products with the same activities and rates of ice crystal formation. Elimination of the NH_4I reduces or eliminates solution corrosion of metals other than stainless steel.

On conclusion of these investigations using acetone solution combustion techniques for studying correlations of chemical composition with rates and mechanisms of ice crystal formation and nucleus activities, no further investigations into nucleant aerosol chemical compositions were conducted by the author until 1998. At this time, the author was asked to assist in the establishment of a facility to manufacture pyrotechnic generators of ice nucleus aerosols for use in the Texas weather modification programs, and to devise suitable compositions for efficient operations.

4. PYROTECHNIC COMPOSITIONS

The generation of AgI containing ice nucleus aerosols by pyrotechnic combustion in the United States was first demonstrated by Thomas J. Henderson, Atmospherics Inc., who used red road

flares to which small amounts of AgI had been added for orographic cloud seeding in the Kings Canyon in the Sierra Nevada mountains of California. (Henderson, 1958). Ice nucleus activity testing during a Yellowstone Field Research Program sponsored by Vincent Schaefer, demonstrated the formation of copious quantities of ice crystals from the aerosol from these flares (T.J. Henderson, private communication). Also in 1958, the discovery that silver iodate, (AgIO_3) could be used as an oxidizer in pyrotechnic formulations to generate AgI aerosols was made and patented (Burkardt et al 1962). On combustion, the AgIO_3 is reduced to AgI , which is vaporized, expelled and recondensed to form the aerosol. The use of AgIO_3 as an oxidizer in pyrotechnics and a source of AgI was exploited by the Naval Weapons Center, China Lake, California. Initially, several varieties of large cast composite pyrotechnic generators were developed for use in Project Stormfury, a hurricane seeding program (Vetter et al, 1970). Subsequently, the program changed over to the development of pressed composite pyrotechnic formulations to satisfy the need for smaller, more efficient generating devices. This change led to the development of the LW-83 (TB-1) composition consisting of 78% AgIO_3 , 12% Al , 4% Mg , and 6% epoxy resin binder (St. Amand et al, 1970). This formulation is still in use, on occasion, today. The fortuitous discovery by Thomas W. Slusher, of Nuclei Engineering Inc., that the ice nucleus activity of the combustion aerosols from the Navy TB-1 composite pyrotechnic could be significantly increased by the addition of small amounts of organic compounds containing chlorine to the pyrotechnic formulation (Slusher, 1978, Sax et al 1978) was an important contribution to ice nucleus aerosol generation technology. This discovery was interpreted to confirm that the structure match of nucleant and ice was improved by the conversion of the AgI crystal structure to an $\text{Ag}(\text{Cl})\text{I}$ species, thus improving the ice nucleus activity.

High AgIO_3 type pyrotechnic compositions that generate active ice nucleus aerosols, that function rapidly by the condensation-freezing mechanism, have apparently not been developed and characterized for field use. To this end, and to compliment the Navy TB-1 and the NEI TB-1 with HCB compositions, the Concho Cartridge, Co. Inc., of San Angelo, Texas can now produce the BF-1 20g. ejectable pyrotechnic, in the 20mm cartridge configuration. This pyrotechnic is formulated to generate, on combustion, an $\text{Ag}(\text{Cl})\text{I}-0.5 \text{ NaCl}$ nucleus aerosol. The ice nucleus activity of this

aerosol, as determined in the isothermal cloud chamber at Colorado State University is 4.98×10^9 and 5.56×10^{10} at -6°C and 2.20 - 6.36×10^{12} . These values compare favorably with those reported for the NEI TB-1 with HCB (Sax et al 1979).

Pyrotechnic flare compositions need to be further improved in order to decrease coagulation, and yield nucleus aerosols with higher efficiencies of aerosol generation, high ice nucleus activity and rapid rates of ice crystal formation. This improvement can be achieved. The Russian silverspare pyrotechnic composition, used in the Swiss hail suppression program Grossversuch IV is a low AgI content flare. On combustion it produces an aerosol with excellent ice nucleus activity at warm temperatures and which functions rapidly by a condensation-freezing mechanism (Federer and Schneider, 1981). A development program to formulate similar high efficiency pyrotechnics is currently underway at the Concho Cartridge Co., Inc. of San Angelo, Texas.

5. DISCUSSION

The research program at Colorado State University's Cloud Simulation and Aerosol Laboratory had benefits beyond the elucidation of the relationships between nucleant chemical compositions, ice nucleus activities, and the rates and mechanisms of ice crystal formation. The emphasis on nucleus aerosol generation by combustion of acetone solutions enabled more rapid progress to be made, than could be achieved by other techniques. The program has led to the wide usage of the Ag(Cl)I-0.5 NaCl nucleus composition, (The Cl to I ratio in the Ag(Cl)I nucleant is approximately 22:78), for field programs. The Kansas hail suppression program and the hail suppression program in France (Dessens et al 1998) use this nucleus aerosol with documented success. The hail suppression programs in North Dakota (Langerud and Moen, 1998) and in Alberta Province, Canada use a modification of this nucleus. The winter orographic cloud seeding program in the Kaweah basin of the Sierra Nevada mountains of California, conducted by Thomas J. Henderson of Atmospherics Inc., uses this nucleus and is accumulating data on its utility relative to that of the AgI contact nucleus from the AgI-NH₄I-acetone-water combustion system.

The identification of AgI and Ag(Cl)I as "contact" nuclei having a rate of ice crystal formation which is a function of the cloud droplet

concentration, was important mechanistically. In hindsight this finding should not have been taken as a recommendation for the use of these nuclei over that of the 2AgI-NaI nuclei, especially for winter orographic cloud seeding. The superior activity of the contact nuclei is counterbalanced by their slow contact rate with cloud droplet concentrations of ~ 200 per cubic centimeter. Contact nuclei can, however, function rapidly to produce ice crystals by exposure to water supersaturations providing that they are generated, together with substantial quantities of water vapor, at temperatures of -6°C and below (Finnegan and Pitter, 1988, DeMott, 1995). This can often be accomplished by locating acetone burning generators at appropriate positions upwind of the target area. The 2AgI-NaI and 2AgI-KI nuclei can also function rapidly on exposure to transient water supersaturations (Blumeinstein, 1987) and should demonstrate rapid ice crystal formation rates on passing through cold cloud bases.

Information gained on new nucleant compositions generated by solution combustion concerning activities, mechanisms and rates of ice crystal formation can be translated to pyrotechnic combustion products. Proper attention must be paid to the pyrotechnic chemistry, to the arts and science of pyrotechnic manufacture, and to the limitations of pyrotechnic combustion for the translation to be successful. One of the difficulties encountered in the use of pyrotechnics is the relatively high rate of aerosol generation from pyrotechnic formulations containing high concentrations of AgIO₃ as the oxidizer. The concentration of ice nucleus particles in the "smoke" or exhaust plume is extraordinarily high, which enhances coagulation or aggregation of the particles. This process has led to a serious misconception concerning the ice nucleus activities of pyrotechnic aerosols in field applications. Coagulation of the aerosol from the high AgIO₃ formulation pyrotechnics, such as the Navy TB-1 and the NEI- TB-1 with hexachlorobenzene, reduces the activity (the number of particles, per gram of nucleant, of 200 to 400 Angstroms diameter which are responsible for ice crystal formation) by about two orders of magnitude. In other words, 99% of the potentially useful nuclei particles are removed by coagulation in the pyrotechnic exhaust. This was shown, in cloud chamber tests, by comparing ice nuclei activities from NEI TB-1 combustion aerosol generated above the wind tunnel fan and the air flow straighteners, with those activities from samples of the aerosol generated below the fan at the entrance to the wind tunnel. The air flow speed for

these tests was ~110 miles per hour or "max fan speed". At -20°C , the activity of the aerosol generated above the fan in laminar flow air was 1×10^{13} IC/gram. The activity of the aerosol generated below the fan was $\sim 1 \times 10^{15}$ IC/gram. Rapid dilution and temperature quench, leading to decreased coagulation can account for the increase in activity of the aerosol generated below the fan (unreported data from the Cloud Simulation and Aerosol Laboratory, Colorado State University). Pyrotechnics burning in free fall or in-place on an aircraft wing might be expected to resemble those burning in the laminar air flow in the CSU wind tunnel. If so, the practice of using high AgIO_3 content burn-in-place pyrotechnics on aircraft to inject large quantities of AgI or $\text{Ag}(\text{Cl})\text{I}$ aerosol for situations requiring such treatment, can be seen to be of little value when coagulation of the aerosol reduces the nucleant output by two orders of magnitude.

Considering this occurrence, together with the fact that different nucleus compositions may function by different mechanisms with different rates and activities, the practice of evaluating field programs or of comparing field programs using only the total weight of silver iodide dispensed per cloud or per season (See for example, Kessler, 1998), should be reconsidered.

Another misconception, still encountered on occasion, is that information from laboratory determinations of activities, and rates and mechanisms of ice crystal formation does not apply to nucleus aerosols used in the "real" atmosphere. Claims are made that nuclei can and will function by sublimation (direct vapor to ice, without the formation of an intermediate liquid phase) to produce the desired results. There is no reason to suspect that heterogeneous ice nuclei will not function by the mechanisms determined in chamber characterizations when generated in the atmosphere. For example, the rapid nucleation, observed when AgI nuclei (of any type) are exposed to transient supersaturations with respect to liquid water in laboratory studies (Blumenstein et al 1987, Feng and Finnegan, 1989) are also observed when AgI aerosols are generated in supercooled fogs from acetone solution combustion (Finnegan and Pitter 1988). Moreover, depositional nucleation, according to N. Fletcher (1962) can occur only on substrates which are completely insoluble in water. Silver iodide has a limited but finite solubility in water of \sim one part in 10^9 parts of water. Wetting AgI particles leads to rapid solution, ionization of the dissolved AgI , and equilibrium formation of surface patches of hydrated Ag^+ and I^- ions on the particles

(de Keizer and Lyklema 1981). This leads to hydrophilicity of the particles and the freezing mechanism described by Finnegan (1998), for immersed particles. Studies (Corrin and Barchet 1970, Zettlemoyer et al 1961, Burstein, 1955, and Tcheurekdjian, et al, 1964) have demonstrated that conventionally prepared AgI contains hygroscopic salts co-precipitated during manufacture, which provide hydrophilic sites for water adsorption from the vapor phase, leading to slow nucleation at -20°C at water saturation in a droplet cloud (DeMott et al 1983, Finnegan, 1998).

There is no published experimental evidence for a depositional nucleation, and one should not invoke one for AgI nucleus aerosols.

6. CONCLUSIONS

Based on the above information, it would appear that weather modification field programs, either exploratory or operational, would best be conducted using heterogeneous ice nucleus aerosols that function rapidly by a condensation followed by freezing ice crystal formation mechanism, have been tested and shown to have high ice nucleus activity, particularly at temperatures above -10°C and a threshold temperature of activation of -5°C or -6°C . These aerosols should be generatable in a consistently reliable and reproducible manner by either solution combustion or pyrotechnic combustion.

The solution combustion generation process for nucleus aerosols with these desired characteristics has already been achieved. The $\text{Ag}(\text{Cl})\text{I}-0.5 \text{NaCl}$ aerosol is in wide use with documented utility. Minor improvements in ice nucleus activity and rate of ice crystal formation can be made by increasing the NaCl to $\text{Ag}(\text{Cl})\text{I}$ ratio but at the expense of increased generator maintenance. No further efforts in solution combustion generation of ice nucleus aerosols are contemplated by this author.

The pyrotechnic combustion process for generating ice nucleus aerosols with similarly desirable characteristics is still in a state of development. There appears to be no unanimity of opinion on chemical composition of pyrotechnic to employ. Many of the formulations are proprietary in nature. Problems of functional inconsistency and reliability have been encountered. It is hoped that continued research, development and scientifically conducted field trials will improve the operational

utility and value of these ice nucleus generator devices.

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