Relation Between Ice-Forming Ability and Conditions of Formation of Silver Iodide Nuclei[1]

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ABSTRACT

An investigation of the silver iodide nuclei produced by a Skyfire generator has revealed the effects of the stoïchiometric ratio and the rate of consumption of AgI on the size spectrum and the ice-forming ability of the nuclei.

A stoïchiometric ratio of 1.5 produces nuclei active at high temperature; as a consequence, it is desirable in cloud modification activities. Acetone vapor on the other hand, seems able to deactivate silver iodide nuclei.

1. Introduction

After many years of empirical work, during which time everyone organizing a weather modification experiment has released the maximum number of nuclei consistent with his funds and techniques, it seems that any further progress on the subject requires a seeding of clouds at a determined point and moment, and with a number and a kind of ice nuclei, selected according to the type of clouds and the desired type of modification.

To attain this, it is necessary to have generators of ice nuclei which produce closely controlled ranges of nuclei types and sizes.

Our work has consisted in reexamining, theoretically and experimentally, the generation of the most frequently used type of nuclei, silver iodide, in order to determine how to control the number, the size and the nature of the resulting nuclei and to adapt them to the stated purpose.

An experimental generator and a wind tunnel (Figs. 1 and 2) have been specially designed and built to do this work (Pejoux, 1970). The generator is of the Skyfire type (Fuquay, 1960), burning an AgI-NaI acetone solution of a known concentration; the supply, with solution, propane and air, is controlled. The apparatus is set in an iron chamber with a chimney in which a nitrogen flow gives a known smoke dilution; the temperature of the flame is measured with a thermocouple. The resulting ice nuclei are sampled at the top of the chimney; their nature is determined by X-ray spectrography and their size with an electron microscope after they have been collected and shadowed on collodion film. Their ice-forming activity is measured in a 10-liter mixing cloud chamber (Soulage, 1964), the ice crystals being counted in a sugar solution or on a Formvar film.

In this way, we simultaneously determined the main characteristics of the nuclei and the parameters governing their generation, and so established relations between them.

With the above equipment, we have mainly investigated four points:

1) The spraying and the combustion of droplets of a silver iodide solution.

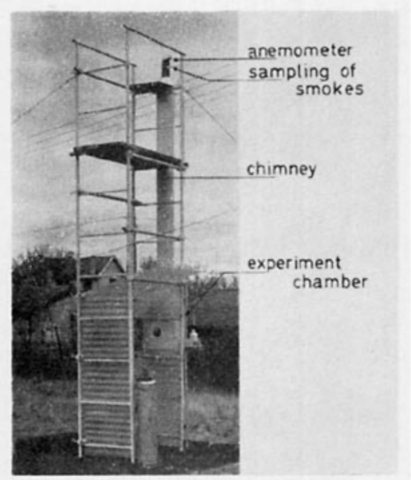

Fig. 1. Silver iodide generator and wind tunnel.

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[1] Presented at the American Meteorological Society Conference on Clouds Physics, 24–27 August 1970, Fort Collins, Colo.

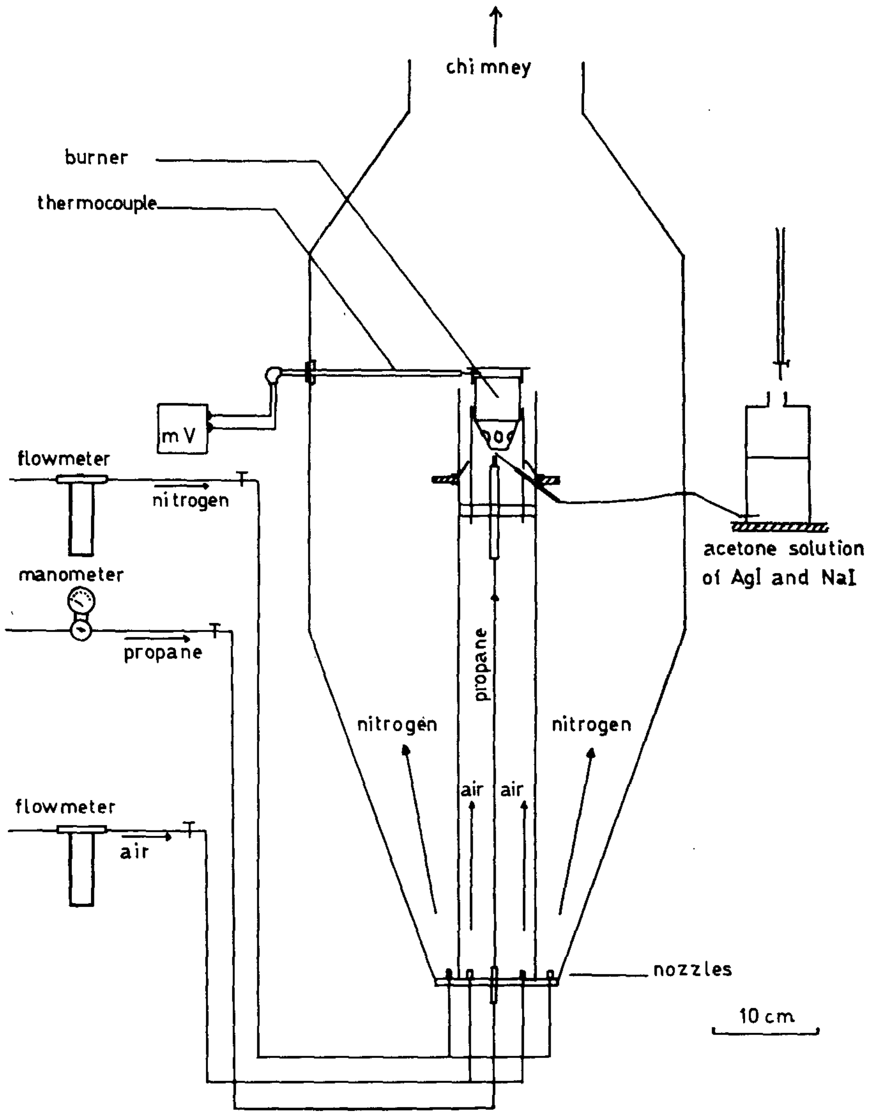

FIG. 2. Schematic diagram of the experiment chamber.

2) The size spectrum of the nuclei produced by this combustion.
3) The ice-forming ability of the nuclei.
4) The poisoning of the nuclei-acetone vapor.

Subsequent efforts were concerned with theoretical explanations of the experimental results.

2. Spraying and combustion of the solution droplets

The volatilization of acetone droplets is instantaneous only if they are smaller than 8 μm in diameter, according to Essenhich and Fells (1960). For larger droplets, according to the same authors, the rate of decrease of the radius r is given by

$$\frac{dr}{dt} = -\frac{1}{r}\frac{\lambda\Delta T/Q\sigma}{1-r/a},$$

in which r is the radius of the droplet, a the radius of the front of combustion around the droplet, λ the thermal conductivity of the gasses around the droplet, Q the vaporization heat of the liquid of the droplet, σ its specific mass, and ΔT the temperature difference between the droplet and the flame.

In this equation, the ratio r/a increases and approaches 1 when the partial pressure of oxygen increases; it follows that it is advantageous to add oxygen to make the combustion easier. On the other hand, dr/dt varies directly with ΔT; hence, the use of an auxiliary combustible which raises the flame temperature is advantageous. Our measurements show that with propane, complete volatilization is possible for droplets $<20\,\mu$m in radius, if the stoïchiometric ratio[2] equals or exceeds 1.

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[2] Ratio of the air actually used to the theoretical amount necessary for complete combustion of the acetone and propane.

Table 1. Size characteristics of nuclei as a function of the concentration of AgI in the acetone solution and the stoïchiometric ratio of combustion in the burner.

AgI concentration in the acetone solution, C (gm liter^{-1})	Stoïchiometric ratio r_s	Nuclei Median radius $\bar{R}$ (Å)	Nuclei Median volume $\bar{v}$ (mμ^3)	Nuclei Number per gram AgI N_t	Nuclei Number per second N_s
1.5	1	250	2.9×10^{-4}	8.4×10^{14}	2.5×10^{10}
	1.5	290	4.4×10^{-4}	5.6×10^{14}	1.7×10^{10}
15	0.5	510	1.5×10^{-3}	1.6×10^{14}	4.8×10^{10}
	1	350	8.5×10^{-4}	2.9×10^{14}	8.7×10^{10}
	1.5	560	2.2×10^{-3}	1.1×10^{14}	3.3×10^{10}
100	1	560	2.8×10^{-3}	8.6×10^{13}	1.7×10^{11}
	1.5	950	5.7×10^{-3}	3.4×10^{13}	6.8×10^{10}

3. Size spectrum of nuclei

For a constant flow of propane and a constant smoke dilution, the size spectrum of the resulting nuclei is mainly a function of the concentration of the AgI-NaI solution and the stoïchiometric ratio.

Table 1 shows how the size spectrum varies as a function of these two factors. We note the following points:

1) For a constant concentration, the radius of the nuclei decreases when the stoïchiometric ratio varies from 0.5 to 1, then increases when this ratio changes from 1 to 1.5.

2) For a constant stoïchiometric ratio, the median radius of the nuclei increases with the concentration of the silver iodide in the acetone solution, while the mean volume also increases with the concentration, but not at the same rate.

These results might be explained as follows. The size of the nuclei depends on the number of embryos formed in a unit volume of AgI vapor, on the partial pressure of this vapor, on the exposure time of each embryo to the vapor, and on the completeness of the vaporization of the silver iodide.

For a constant concentration, when the stoïchiometric ratio increases from 1 to 1.5, the thermal gradient in the flame and, as a consequence, the supersaturation of the AgI vapor, decrease; this results in fewer embryos and larger nuclei for the same quantity of silver iodide. When the stoïchiometric ratio decreases from 1 to 0.5, another phenomenon occurs: the combustion of the droplets becomes less efficient and the solution droplets are not completely vaporized; thus, the median radius of the nuclei increases, as was observed.

For a constant stoïchiometric ratio, when the concentration of the solution increases, the supersaturation of AgI vapor increases; this results in both an increase of the number of embryos in a unit volume of vapor (and, as a consequence, an increase of the output per second) and an increase of the growth rate of the embryos (and, therefore, an increase of the radius of the nuclei). This explains why the mean volume of the nuclei does not increase in the same ratio as the concentration and why the output per gram of silver iodide certainly decreases, but at a lower rate than if the AgI vapor were distributed on the same number of embryos per unit volume.

4. Ice-forming ability of the nuclei

Fig. 3 shows the ice-forming ability of the nuclei, the size variation of which has just been discussed. It shows the number of nuclei per gram of AgI effective at −20C, −8C, and above −6C as a function of the stoïchiometric ratio, and for a concentration of the AgI-NaI solution equal to 15 gm liter^{-1}.

We note that the curves for −20C and −8C exhibit a maximum for a stoïchiometric ratio of 1, in agreement with the earlier work of Steele and Sciacca (1966). This maximum coincides with the maximum of the total number of nuclei and with the minimum of the

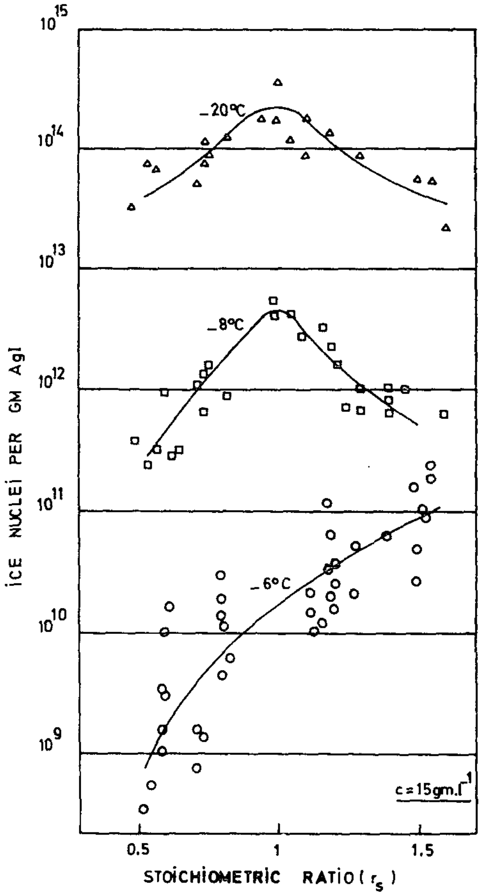

Fig. 3. Variation of the number of ice nuclei per gram of AgI with the stoïchiometric ratio of combustion in the burner for different cloud temperatures and an AgI concentration of 15 gm liter^{-1}.

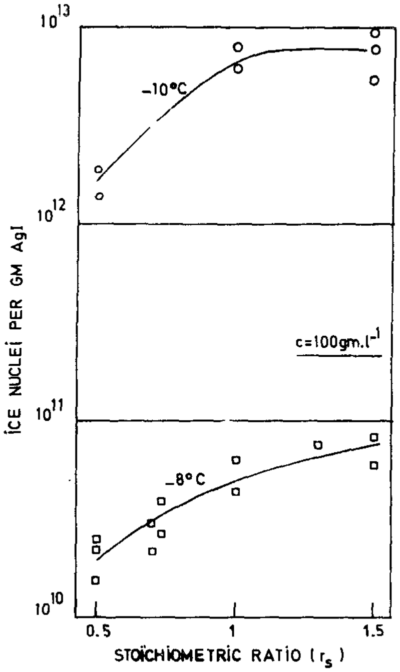

FIG. 4. As in Fig. 3 except for an AgI concentration of 100 gm liter^{-1}.

observed previously median radius for the same stoïchiometric ratio of 1.

The curve for −6C exhibits a different feature. Instead of decreasing, the number of nuclei increases as the stoïchiometric ratio increases from 1 to 1.5. This result is not inconsistent with the above. The increase of the number of effective nuclei, notwithstanding the decrease of the total number of nuclei, may be explained by the increase of their size which enables a greater part of them to be effective at high temperatures when $r_s = 1.5$.

The above increase does not occur when r_s changes from 1 to 0.5, in spite of a similar increase of the size of the nuclei; this may be due to a loss of nuclei owing to an incomplete vaporization of AgI and to the poisoning by acetone vapors that we shall study in the next section.

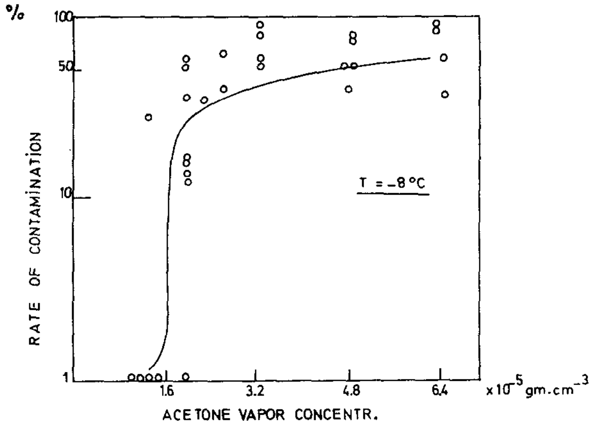

FIG. 5. Variation of the rate of contamination of ice nuclei with the acetone vapor concentration.

Likewise, when the concentration of the solution is changed from 15 to 100 gm liter^{-1} (Fig. 4) and when, as a consequence, we again have larger nuclei, the number of effective nuclei is not as significant a function of the stoïchiometric ratio.

These results confirm the part played by the size of the nuclei in their ice-forming ability.

The results are important for weather modification trials requiring nuclei effective at high temperature (hail prevention, for instance), since they suggest methods of producing a maximum of nuclei efficient for such modification.

5. Poisoning of AgI nuclei by acetone vapor

This poisoning was revealed by the experiments with a stoïchiometric ratio of 0.5. The results are given in Fig. 5, which shows the percent of deactivated nuclei as a function of the concentration of the acetone in the smokes. The measurement was made by comparing two samples of nuclei produced by our generator, introduced into identical glass flasks, one contaminated by acetone, the other without contamination. We see that an important poisoning intervenes when the acetone concentration exceeds 1.6×10^{-5} gm cm^{-3} in the smokes.

For a stoïchiometric ratio of 0.5 we observed 2×10^{-5} gm cm^{-3} of acetone in the smokes of our generator, which may explain the result given above.

6. Theoretical interpretation

The relation between size and ice-forming ability of the nuclei has been examined from two points of view: the kinetic theory of nucleation (Fletcher, 1959, 1968; Isaka, 1966a, b, 1969) and the "site" theory of Katz (1962).

The first introduces the notion of a temperature threshold for the nuclei (the surface of which is considered spherical and homogeneous) as a function of their size; the second introduces the notion of nucleation sites distributed at random on the surface of the nuclei, the presence of which is necessary to the activity of a particle.

In order to test the first of the theories, we calculated the theoretical number of ice nuclei corresponding to the observed nuclei size and compared them to the number of ice nuclei determined experimentally. We made this comparison, on the one hand, for AgI-NaI nuclei taking into account the temperature threshold measured by de Pena (1964), and on the other hand, for pure AgI nuclei, considering the threshold measured by Mossop and Tuck-Lee (1968) and ourselves. The results are shown in Fig. 6, where the ordinate is the ratio between the theoretical and experimental number of nuclei, and the abscissa the temperature. We note a qualita-

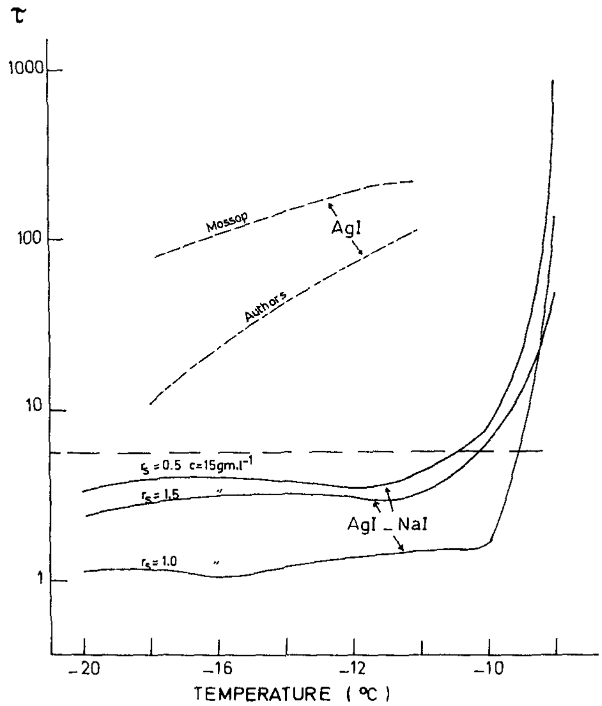

FIG. 6. Variation of the ratio τ between the number of ice nuclei calculated and the number of ice nuclei determined by experiments with temperature for various stoïchiometric ratios.

tive agreement between the two numbers (theoretical and experimental) for the AgI-NaI nuclei and for temperatures < -8C. This agreement is not observed for the same nuclei at temperatures > -8C. Likewise, it is not observed for pure AgI nuclei, the calculated number being from 10 to 100 times higher than the measured number.[3]

In the same way, to test the second theory ("site" theory), we calculated the size spectra of the nuclei effective at various temperatures, and obtained almost the same spectra, whatever the temperature. This result is inconsistent with our experiments (Pejoux, 1970).

Thus, neither of the two theories thoroughly explains our experimental results, although the first seems to be more useful.

Acknowledgments. This paper uses, as its basis, the dissertation of one of the authors (R. Pejoux) submitted to the University of Clermont-Ferrand for the degree of Doctor of University. We are indebted to Profs. Bricard, Bernard and Hitschfeld for the supervision of this dissertation.

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[3] It seems as if the hygroscopicity of AgI-NaI nuclei (which is not part of the kinetic theory of nucleation, however) tends to bring the real activity of the nuclei near their theoretical activity.