A Supersonic Expansion Method of Ice Nuclei Generation for Weather Modification[1]

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ABSTRACT

Aerosols of organic ice nucleating agents were formed by rapid adiabatic expansion, by passing superheated steam laden with the organic vapors through a supersonic nozzle. The effect of the Mach number of the flow through the nozzle on the aerosol particle size and number concentration was investigated. It was found that the number of particles formed increased by a factor of 10^3 as the Mach number increased from 1 to 2, but further increases in Mach number had little effect. The ice nuclei activity spectra of the organic aerosols thus generated showed plateaus at colder temperatures, with approximately 10^{14} nuclei per gram below -9°C for 1,5-dihydroxynaphthalene and 10^{12} g^{-1} below -7°C for metaldehyde.

Coagulation rates of both organic smokes were determined. Coagulation of 1,5-dihydroxynaphthalene aerosols followed the Smoluchowski equation. Coagulation of metaldehyde aerosols was not only extremely rapid, but in addition the coagulation constant increased with time. The latter effect was attributed to the strong electrical dipole of metaldehyde particles.

1. Introduction

From the beginning of modern ice phase weather modification, two types of ice nucleants have been widely used. The first are evaporative coolants like dry ice and liquid propane. The ice nucleation mechanism of the coolants is homogeneous freezing of droplets formed by evaporative cooling. Nucleants of the other type are crystalline solids dispersed in aerosol form, typically represented by AgI. Coolants normally form about 10^{12} ice crystals per gram (Fukuta, 1965, 1972; Fukuta *et al.*, 1970, 1971; Hicks and Vali, 1973) independent of the supercooled fog temperature up to almost 0°C. On the other hand, the number of active nuclei per gram of heterogeneous nucleant increases as the fog temperature decreases, and eventually exceeds that of the coolants. Heterogeneous nucleants have been more popular for cloud seeding because of their solid crystalline nature at temperatures above 0°C, the ease of material storage, the availability of practical smoke generation techniques, and the larger number of ice nuclei per gram of compound at moderately low temperatures where the rate of phase change of the supercooled cloud into ice is high. As we have pointed out (Fukuta, 1974), the solid form of the smoke particles permits us to disperse the nucleant in the warm or dry air outside the cloud for seeding purposes. In such an air mass, of course, ice crystals generated by coolants cannot survive. Seeding there is often necessary in order to gain time for diffusion of the particles or to avoid use of a costly aircraft with high altitude capability.

Organic ice nuclei belong to the category of heterogeneous nucleants. Their ice nucleabilities, as judged from the nucleation thresholds, are as good as or better than that of AgI. They exhibit some very important features. First of all, prices of the three organic nucleants considered to be suitable for practical use [1,5-dihydroxynaphthalene (DN) (Fukuta, 1966), metaldehyde (MA) (Fukuta, 1963) and phloroglucinol (PG) (Bashkirova and Krasikov, 1957)] are about 10^{-1}–10^{-2} that of AgI. The price of silver has recently risen by a factor of 3. The long-term availability is a serious problem for AgI but not for the organic nuclei compounds. Second, the organics are biodegradable; our laboratory tests with bacteria, algae and fish demonstrated the biodegradability of the organic compounds, but the same tests with AgI revealed that guppies could not survive in a saturated solution, even though very dilute, and their reproductive cycle was harmed (Church *et al.*, 1975), suggesting another severe disadvantage for its long-term and widely spread use. The third feature is that organics provide little or no possibility of giving downwind effects due to persistence of nuclei in the atmosphere. The only disadvantage known with the organics was the slightly low number of active nuclei per gram compared with that of AgI even if the condensation method of jet vapor mixing

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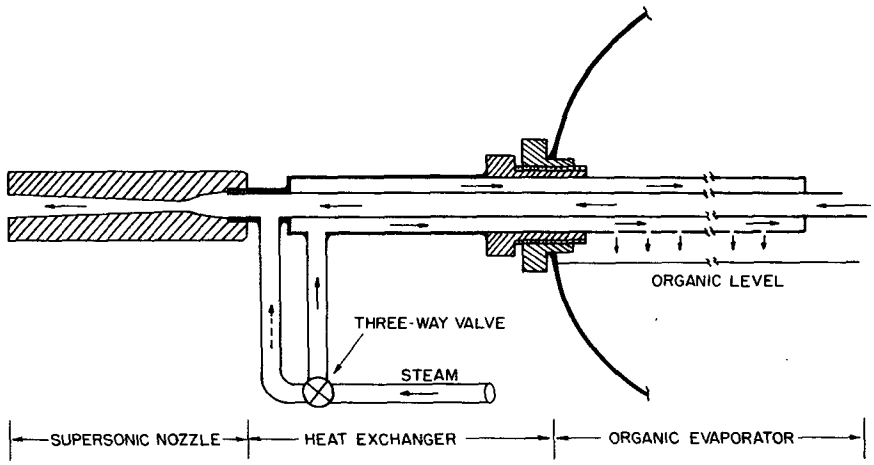

FIG. 1. Schematic drawing of the nozzle adaptor.

was used for nuclei generation. Jet mixing is the most effective method so far applied to the ice nuclei particle generation problem (Fukuta *et al.*, 1966; Fukuta, 1972).

In the jet mixing method, a hot vapor or vapor-laden gas is blown out of a proper nozzle. As the hot vapor mixes with the cold ambient air, it is cooled and becomes supersaturated. To attain a high supersaturation, however, this technique has the following two intrinsic disadvantages. Since quenching of the hot vapor is performed by mixing with cold air, the process simultaneously results in dilution of the vapor, thereby diminishing the supersaturation-increasing effect. Another disadvantage comes because the cooling rate is limited by thermal diffusion across the molecular diffusion boundaries created by the micro-eddies in the mixing region. In such a supersaturation field formed by the simultaneous diffusion of heat and vapor into the quenching air, homogeneous condensation takes place. As the rate of homogeneous nucleation is directly related to the true or effective supersaturation, which is a function of the cooling rate, achievement of an extremely fast cooling rate without vapor dilution is the key to the generation of a large number of smoke particles. The method which satisfies the above requirements must therefore utilize fast internal cooling. For this reason, the supersonic expansion method—a method known to give nearly adiabatic and isentropic cooling due to the fast and continuous expansion—has been introduced into the technology for organic ice nuclei smoke generation (Fukuta, 1972; Paik *et al.*, 1972). This paper reports results of our recent study on the supersonic nozzle method using DN and MA as ice nuclei smoke-producing materials.

2. The organic ice nuclei generator

An organic ice nuclei generator for ground use, utilizing the isentropic or supersonic expansion method of smoke generation, has been developed and tested. Details of the generator have been given elsewhere (Fukuta, 1972). It consists of three major parts: steam generator, organic evaporator, and supersonic nozzle and heater. The steam generator contains about 16 liters of deionized water. Steam pressure is automatically controlled at a set level by means of a pressure sensitive valve, which adjusts the fuel gas flow to the burner. The steam, made hot and dry with the help of a superheating coil, is introduced into the evaporator, which contains about 2 kg of organics in powder form, through small nozzles directed onto the surface of the organic substance. The organic evaporates into the steam, and the vapor-laden steam is then carried to the nozzle via a heat exchanger which is designed to avoid deposition of organic and water in the nozzle. The heat exchanger also allows for preheating and cleaning of the nozzle by simply passing hot steam outside (see Fig. 1).

The temperature of the evaporator was controlled so as not to exceed the thermal decomposition point of the organics. Optimum evaporator temperatures were, respectively, about 200°C for DN and 140°C for MA. Steam as a carrier gas of organic vapor has the advantages of being nonoxidizing, nontoxic and available anywhere. The supersonic nozzles used were designed to have flow capacities of about 2 ℓ s^{-1} in the standard state (see Fig. 2).

3. Cooling during the adiabatic expansion through a supersonic nozzle

To obtain a rapid adiabatic cooling, we utilize the diverging section of the de Laval nozzle (Courant and

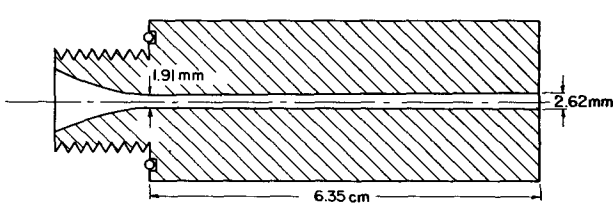

FIG. 2. Design of a supersonic nozzle with Mach number 2.04.

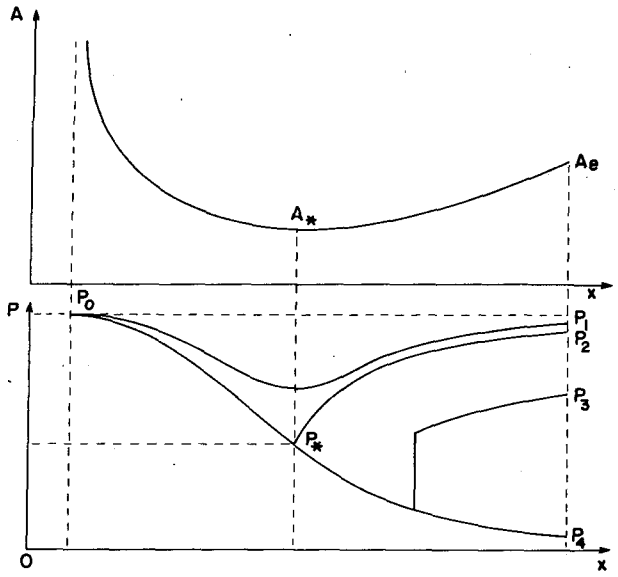

FIG. 3. The pressure P as a function of the position x along the axis of a supersonic nozzle for various flows resulting from various exit pressures (after Courant and Friedrichs, 1963). A is the cross-sectional area of the nozzle. For flow behaviors corresponding to different exit pressures, see text.

Friedrichs, 1963). The de Laval nozzle consists of a converging "entry" section and a diverging "exhaust" section with a "throat" in between. When a high-pressure gas at rest in a stagnation chamber flows through such a nozzle, two different situations arise. Fig. 3 illustrates the nozzle gas flow under different conditions. If the ratio of pressures between the exit and the entry section is above a certain "critical" value (P_2/P_0), the flow, after being expanded in the converging section, is compressed in the diverging exhaust section and the gas flow remains subsonic. When this pressure ratio becomes lower than the critical value, on the other hand, the flow becomes supersonic after passing through the throat, but at a certain place in the diverging section of the nozzle, a shock front forms, the pressure rises and the flow again becomes subsonic. As the pressure ratio is further reduced, the shock front moves toward the exit. At the pressure ratio P_4/P_0, the shock front just reaches the exit, and at any smaller pressure ratio the nozzle gas becomes completely supersonic after passing the throat and expands. The expansion of the nozzle gas entirely controls the cooling.

The flow of a gas, assumed to be noncondensable, is governed by the continuity equation

$$A\rho u=G, \tag{1}$$

where A, ρ, u and G are the cross-sectional area of the nozzle, the gas density, the flow velocity, and the mass flow across this area per unit time, respectively. The adiabatic relation is given by

$$P\rho^{-\gamma}=\text{constant}, \tag{2}$$

where P and γ are the pressure and the adiabatic exponent, and Bernoulli's law is

$$\frac{\gamma-1}{\gamma+1}u^2+\frac{2}{\gamma+1}c^2=c_*^2, \tag{3}$$

where

$$c^2=\frac{\gamma P}{\rho} \tag{4}$$

is the sound speed and c_* is the "critical speed" where the sound speed and the flow speed agree; γ is defined as

$$\gamma=c_p/c_v=1+R/c_v, \tag{5}$$

where R, c_p and c_v are the specific gas constant and the specific heats at constant pressure and constant volume, respectively.

With a sufficiently large pressure ratio between the entry section and the exit, the change from subsonic to supersonic flow takes place at the throat, i.e., $u=u_*=c_*$ and $A=A_*$. Then, from (1)–(4), the Mach number $M=u/c$ of the flow can be correlated solely with the area ratio, as

$$\frac{A}{A_*}=\frac{1}{M}\left[\frac{(\gamma-1)M^2+2}{\gamma+1}\right]^{(\gamma+1)/[2(\gamma-1)]} \tag{6}$$

Using (2) and the ideal gas law

$$Pv=RT, \tag{7}$$

where

$$v=1/\rho \tag{8}$$

is the specific volume, the temperature change associated with this adiabatic expansion may be derived as

$$\frac{T}{T_0}=\left(\frac{P}{P_0}\right)^{[1-(1/\gamma)]}, \quad \frac{P}{P_0}=\left(\frac{\rho}{\rho_0}\right)^{\gamma}, \tag{9}$$

where subscript 0 refers to the stagnation condition of the gas before entering into the converging section.

The first law of thermodynamics for the gas flow states

$$\Delta h=-\frac{u^2}{2}, \tag{10}$$

where Δh is the change in enthalpy per unit mass, which can be expressed for an ideal gas as

$$\Delta h=c_p\Delta T. \tag{11}$$

Then, setting $\Delta T=T-T_0$, and inserting (5) and (10) into (11) to eliminate c_p, we have

$$u^2=\frac{2\gamma}{\gamma-1}R(T_0-T), \tag{12}$$

TABLE 1. Characteristics of supersonic nozzles used in the experiment.[1]

Nozzle Mach number	Dimensions (cm) l	d_*	d_e	T_e/T_0	P_e/P_0	A_e/A_*	P_0 (bars)
1.57	1.08	0.241	0.269	0.776	0.323	1.25	2.61
1.73	3.49	0.241	0.287	0.698	0.190	1.42	4.44
1.81	6.35	0.052 (height) 0.660 (width	0.056 0.912	0.682	0.167	1.51	5.05
2.04	6.35	0.191	0.262	0.636	0.114	1.89	7.40
2.23	4.87	0.155	0.237	0.605	0.080	2.32	10.58

[1] l and d are length and diameter of nozzles. Subscripts e, $*$ and 0 stand for exit, throat and stagnation chamber, respectively. Nozzle holes are all circular except one with Mach number 1.81 which has a flat shape. $P_e \doteqdot 0.843$ bar in Denver. The Mach number and T_e/T_0 are nominal.

or

$$\frac{T_0}{T}=\frac{(\gamma-1)u^2}{2\gamma RT}. \tag{13}$$

Inserting (4), (7) and (8) into (13) yields the important relationship

$$\frac{T_0}{T}=1+\frac{\gamma-1}{2}M^2. \tag{14}$$

Introduction of (9) into (14) results in corresponding expressions for pressure and density ratios:

$$\frac{P_0}{P}=\left(1+\frac{\gamma-1}{2}M^2\right)^{\gamma/(\gamma-1)}, \tag{15}$$

$$\frac{\rho_0}{\rho}=\left(1+\frac{\gamma-1}{2}M^2\right)^{1/(\gamma-1)}. \tag{16}$$

Table 1 shows characteristics of various supersonic nozzles used in this experiment. Since the carrier gas used water vapor, which is condensable, values of M and T_e/T_0 are nominal, where the subscript e denotes conditions at the nozzle exit.

In our experiment, the adiabatically cooling carrier gas, i.e., water vapor, serves as a medium in which nucleation of organic smoke particles from the vapor takes place. The smoke particle formation in the gas stream is controlled by the interaction between the changing condition of the carrier gas and the nucleation-growth process. The carrier gas provides the conditions suitable for nucleation, which is immediately followed by growth of nucleated particles. While the system continues to nucleate and grow particles simultaneously, during the growth process the particles exert a "back pressure" to the condition necessary for nucleation, by abstracting the supersaturated vapor and adding heat, both acting in the direction of reducing the supersaturation. Since nucleation and growth are rate processes, what affects these two processes is the rate of supersaturation generation in the carrier gas, and it is a function of the cooling rate. We therefore estimate the cooling rate in the supersonic nozzle process.

Suppose that, during a short time period dt, the speed of the carrier gas changes by du and the temperature changes by dT. From (2), (7) and (8),

$$\frac{dT}{T}=(1-\gamma)\frac{dv}{v}. \tag{17}$$

From (1) and (8), the total differential of v may be expressed as

$$dv=\frac{1}{G}(A\,du+u\,dA). \tag{18}$$

Then substituting (17) into (18), dividing both sides with dt, using $u=dx/dt$, and rearranging, we obtain

$$\frac{dT}{dt}=(1-\gamma)T\left[\frac{du}{dx}+u\left(\frac{1}{A}\frac{dT}{dx}\right)\right], \tag{19}$$

where x is the distance in the direction of gas flow at the middle of the nozzle. Thus, (19) shows how the cooling of the carrier gas is influenced by the flow velocity and the nozzle shape $(1/A)(dA/dx)$, providing a guide line for nozzle design, although a refinement due to the nucleation and growth interaction for both water and organic vapors will be required in the future.

4. The smoke generator performance test

Smoke produced from the generator was immediately diluted by introducing it into a wind tunnel, which consists of a steel tube 62 cm in diameter, 330 cm long, and a variable speed electric fan. Under typical conditions the dilution ratio for air to the smoke particle-laden steam was 377. To determine the particle concentration, the dilute smoke was transferred to an ultramicroscope cell by means of a syringe. By taking a succession of 30 snap counts and averaging, the number per unit volume was readily deduced from the magnification and dilution factors. Smoke particles always exhibited strong Brownian motion in the ultramicroscopic field, and were very uniform in size. The average radius of DN smoke particles was smaller than 0.1 μm, while that for MA smoke was approximately 0.2 μm.

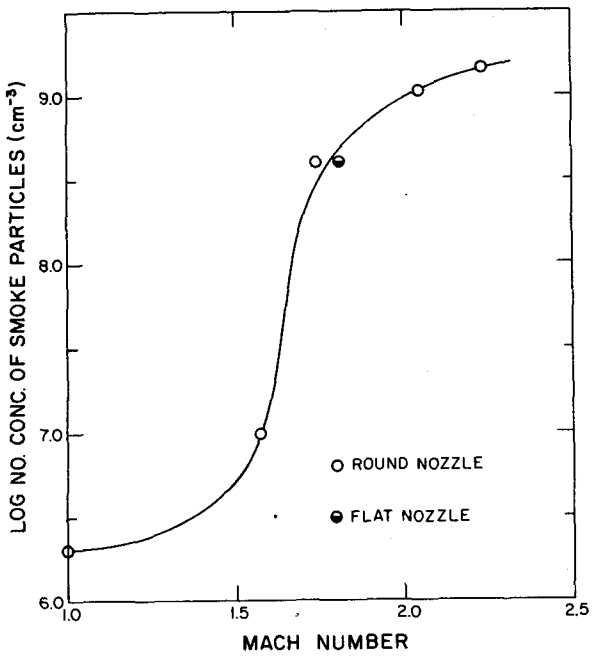

FIG. 4. Number concentration of 1,5-dihydroxynaphthalene smoke in the nozzle gas at the exit under STP with varying Mach numbers. The number concentration was determined by ultramicroscope method.

The ice nucleabilities of both smokes were measured using a University of Denver standard mixing cloud chamber with sugar or polyvinyl alcohol solution to detect ice crystals. In the mixing chamber a hot water reservoir at the top permits fog droplets to form and settle through the chilled air. The number of ice crystals formed on the solution was counted for 5 min by the naked eye. The number of active ice nuclei per unit weight of organics was estimated from the volume of the sample air injected and the total weight of organic particles in it. The weight is deduced from the dilution ratio and the mass flow rate of the organic compound.

For determinations of coagulation rates of organic smokes, the nozzle gas was directly received in a cardboard drum 53 cm diameter, 66 cm high. The coagulation rate was determined by measuring the concentration change of smoke particles with time, using the ultramicroscope.

5. Results and discussion

a. Generation of ice nuclei particles

The smoke generator performance tests were made using nozzles with different flow speeds. Number concentrations of DN smoke particles determined by the ultramicroscope method are shown in Fig. 4. With increasing nozzle gas speed, the smoke concentration increases abruptly. At Mach number 1, the concentration in the nozzle gas at the exit was 2.1×10^6 cm^{-3}; the highest concentration was 1.5×10^9 cm^{-3} at Mach number 2.23, which was the fastest nozzle gas speed used in this investigation. It is clear from the figure that there is a supersonic nozzle condition which increases the smoke particle concentration at least by a factor of 10^3 over that of the sonic mixing process. At higher Mach numbers with the present nozzle design, the smoke concentration approaches a limiting value. This may be attributed partly to the condensation shock developed in the nozzle and partly to coagulation. However, the main restriction appears to be attributed to the rapid reduction of supersaturation by growing particles (Wegener, 1954; Pouring, 1965).

In the adiabatically expanding gas environment where the supersaturation tends to increase, the growing particles already nucleated act against the increase by abstracting moisture and adding heat. This eventually leads the system to a maximum supersaturation which controls the total number of nuclei that are formed during the process. The maximum supersaturation, however, is a function of the cooling rate of the gas environment (Fukuta, 1972). In this regard, although the Mach number used in the present experiment is a direct measure of the temperature the system achieved, it must be regarded only as an indirect measure for the cooling rate. As can be seen in (19), the effect of the cooling rate in relation with the final temperature the system achieves can be much more closely examined by changing the nozzle design. Nevertheless, the results shown in Fig. 4 present high promise for the practical application of the supersonic expansion method.

After the supersonic expansion process where the nozzle gas could mix with the ambient air, it was thought that the mixing process might induce additional nucleation of particles when the ambient air temperature was low, and allow already nucleated particles to evaporate if the temperature had been high. To observe the effect of mixing the nozzle gas with air on smoke particle formation, a series of experiments was made at different ambient temperatures ranging from -15 to $+20$°C. There has also been some doubt about possible coagulation loss of the formed particles in the nozzle gas during the process of mixing and diluting with the ambient air. Therefore, a flat supersonic nozzle which creates faster mixing was made and tested under similar experimental conditions. The mixing process did not appear to affect the population of particles after the supersonic process.

The measured ice nucleabilities of DN and MA smokes are shown in Fig. 5, compared with that of silver iodide produced by an airborne AgI-NaI acetone burner (Smith *et al.*, 1966). The number of ice crystals produced by DN smoke rapidly reaches a plateau value, approximately 10^{14} g^{-1} at -9°C, while MA smoke more gradually approaches a value at 10^{13} g^{-1} at -7°C. The nucleability of MA smoke produced by jet mixing (Fukuta *et al.*, 1966) was less by a factor of 10^2 at the

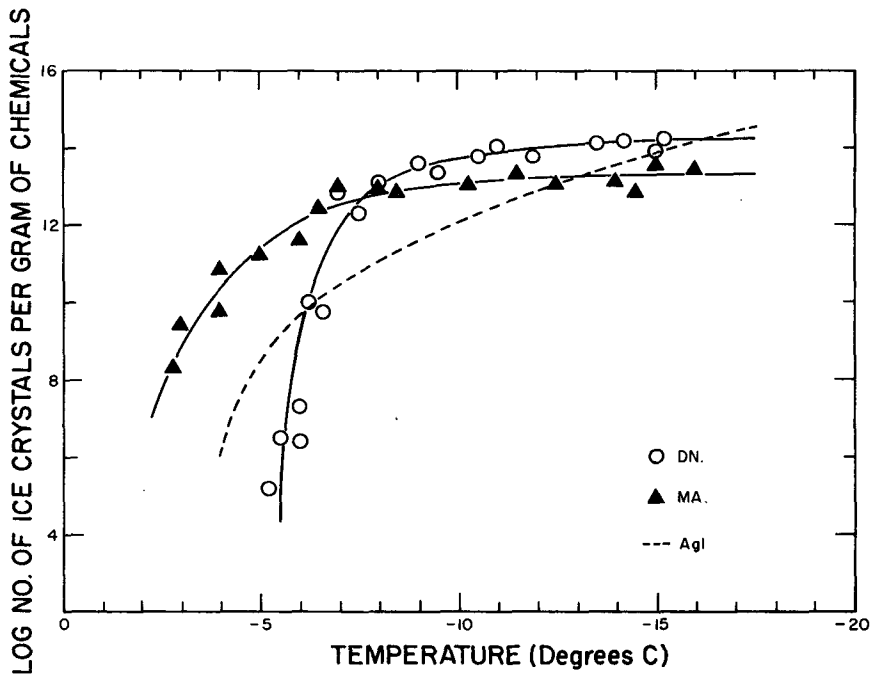

FIG. 5. Ice-nucleability of 1,5-dihydroxynaphthalene (DN) and metaldehyde (MA) generated using the Mach number $M=2.04$ nozzle with $P_0=7.4$ bar. A mixing cloud chamber was used for nucleation test.

same temperature. Results imply that at relatively warm temperatures organic nuclei prepared with the present method are much more active than the silver iodide nuclei, and their activity spectra are independent of temperature in the plateaus.

b. Coagulation of ice nuclei smokes

Since the ice nuclei can be lost due to coagulation during the process of smoke particle dispersion on their way to reach the ice nucleation zone in a cloud, an attempt was made to determine coagulation rates of DN and MA smokes. The rate of smoke particle coagulation can normally be described by the Smoluchowski equation

$$\frac{1}{n}-\frac{1}{n_0}=Kt, \tag{20}$$

where n_0 and n are particle concentrations at time $t=0$ and t, and K is the coagulation constant which depends on the particle size. Coagulation rates of DN smoke measured using the ultramicroscope are shown in Fig. 6. As can be seen, the rate was found to follow the Smoluchowski equation very closely, regardless of the initial smoke concentration. The coagulation constant K was found to be 4.2×10^{-9} cm^3 s^{-1} at 13°C, which is about ten times larger than that of normal oleic acid smoke particles (Green and Lane, 1964) and is comparable to that of 0.01 μm radius particle (Fuchs, 1964), suggesting small sizes for the produced DN smoke particles.

Coagulation rates of MA smokes are also shown for four different initial concentrations in Fig. 7. Here

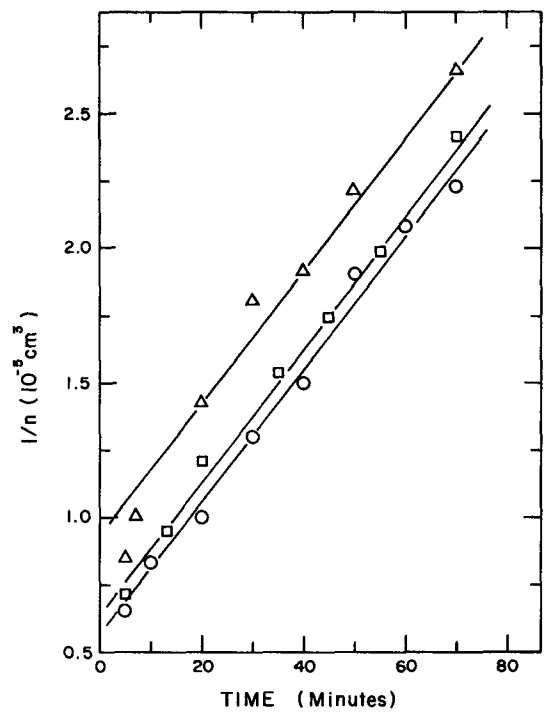

FIG. 6. Reciprocal of partical concentration versus time for 1,5-dihydroxynaphthalene smoke at 13°C. The $M=2.04$ nozzle was used for smoke generation and the number of particles was measured by means of ultramicroscope.

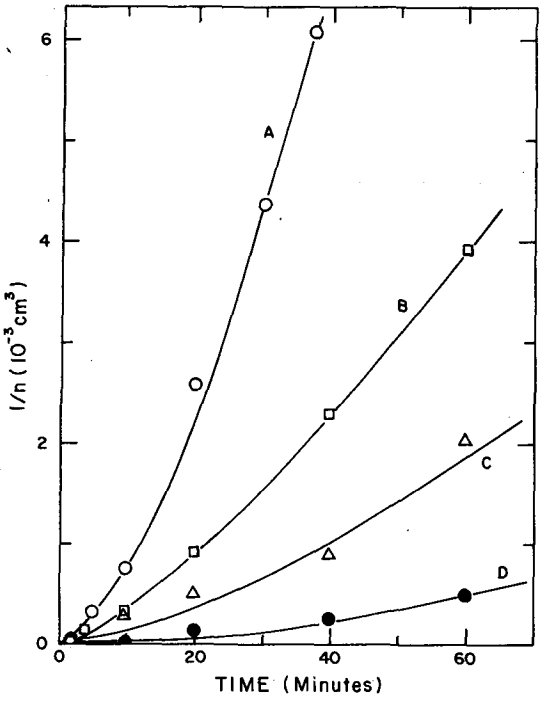

FIG. 7. As in Fig. 6 except for metaldehyde smoke at 13°C.

the initial concentration increases in the alphabetical order of the labels on the curves. The coagulation of MA smoke is very fast, and the rate of coagulation increases as the concentration decreases. From rate curves, the coagulation constant K of MA may be expressed in the simple form

$$K=K_0 n^{-\frac{1}{2}}, \tag{21}$$

where K_0 indicates the coagulation constant at $n=1$ cm^{-3} and is a function of the initial polydispersity of MA smoke. In the present experiment, values of $\log K_0$ varies from -4.6 to -5.6. The higher the polydispersity, the larger were the observed $\log K_0$ values. Gillespie (1953) also reported a concentration-dependent coagulation rate for charged Vycor aerosols. Such a unique behavior of MA smoke may be attributed to the strong dipolar property of the crystal and the formation of a polydisperse aerosol in the course of coagulation, which may lead to changes in the charge distribution. This result quantitatively supports our earlier finding about the coagulation rate (Fukuta, 1963) and is in good accordance with our recent observation that MA particles are dominant contact ice nuclei.

6. Conclusions

For ice nuclei generation, the supersonic expansion was found to be a highly efficient method, particularly for organic ice nucleus compounds. The principal findings from the present experiment may be summarized as follows:

1) The condensation of organic vapor in a supersonic nozzle produces monodispersed smoke particles. Particle radii were less than 0.1 μm for DN and less than 0.2 μm for MA.

2) The supersonic expansion process plays the dominant role for ice nuclei particle production, and the subsequent mixing with the ambient air hardly affects the population of particles.

3) With nozzle gas speed increasing from Mach number 1 to 2, the smoke concentration increases abruptly by a factor of about 10^3.

4) At relatively warm temperatures, organic ice nuclei prepared by the present method are more active than silver iodide nuclei. Their activity spectra have plateau values at lower temperatures: about 10^{14} g^{-1} below -9°C for DN and about 10^{13} g^{-1} below -7°C for MA.

5) The coagulation of DN smoke follows the Smoluchowski equation very closely with the coagulation constant 4.2×10^{-9} cm^3 s^{-1}, whereas the coagulation of metaldehyde smoke is faster and depends largely on its concentration and polydispersity. The higher the smoke concentration, the slower was the observed coagulation.

It is recommended that further investigation on the ice nucleant smoke production process using the supersonic expansion method be carried out by changing the nozzle contour design as well as the vapor-laden gas condition, in order to control the nucleation-growth interaction of ice nucleant particles, the key to the problem.

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