Effect of Time on the Properties of Crystallization Agents: Ice-forming Aerosols

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Abstract **– The results of studies of the efficiency (yield) of pyrotechnic compositions used for the prevention of hail in the Republic of Moldova as a function of storage time are described. The studies are performed using an ingenious installation based on a small aerodynamic stand, which makes it possible to control the efficiency of ice-formation of full-size generators in the on-line mode and to study the nucleation activity of new reagents under dynamic conditions. It is shown that the efficiency of the pyrotechnic compositions based on silver iodide (AgI) decreases depending on storage time. At the same time, the decrease in yield for given standard storage conditions is on the order of 10% per year.**

Index Terms **– Ice-forming aerosol, silver iodide, AgI, aerodynamic stand**

I. INTRODUCTION

The relevance of work on active impacts for the prevention of hail, the redistribution of precipitation, and the thinning of fog and clouds does not give rise to doubts. These works are carried out in many states. In the RM, large-scale works on the protection of agricultural crops from hail damage are carried out on a regular basis (in 2010 the area of territories under protection was 1.4 mln ha); thousands of antihail rockets are utilized for these purposes every year.

Along with cooling agents, ice-forming aerosols are widely used as artificial means of crystallization in the practice of active influences on supercooled cloud environments. Ice-forming aerosols are fine particles of a substance that, under given thermodynamic conditions in a cloud, can act as nuclei (seed) for the formation and growth of ice crystals.

Ice-forming reagents are most commonly used owning to a number of technical advantages, which result from the conditions of storage, delivery and dispersion in the zone of cloud seeding, etc. In the case of using these substances, the method of active impacts is based on the formation of a necessary concentration of artificial ice crystals in the seeded part of the cloud environment; that is, in a potentially hail-hazardous cloud environment, it is necessary to form a certain number of active ice-forming nuclei under given conditions in the cloud and known conditions of introduction of reagents into the atmosphere.

Ice-forming reagents are effective means of artificial crystallization of supercooled clouds and fog, which determines their extensive use in the practice of active influences (AIs) in many countries, including the Republic of Moldova (RM). The ice-forming aerosol based on applied reagents is dispersed into cloudy atmosphere by means of special generators (rockets, pyrocartridges, ground-based and airborne devices); their application is determined by techniques and objectives of AIs [1].

To date, we know a wide class of materials whose fine aerosols can initiate the formation of ice crystals with a high probability. One of the most widely used reagents are those based on silver iodide (AgI). Ice-forming pyrotechnical compositions based on silver iodide (AgI), which are used in antihail rockets for impacts on hail-hazardous clouds in the RM, yield more than 10^{13} g^{-1} of active ice-forming particles at a temperature of the cloud (simulated) environment of -10°C.

The theoretical foundation for describing the effects of fine aerosols of ice-forming reagents based on the theory of nucleation of ice on the surface of aerosol particles developed by H. Fletcher [2, 3].

The use of pyrotechnic generators in different exposure techniques requires reliable knowledge of their efficiency. Therefore, before using these tools in the active intervention, it is necessary to experimentally prove their efficiency.

We can state that the result of this impact on a potentially hail-hazardous cloud with ice-forming reagents depends on many factors and, not least of all, on the yield of a particular generator, which depends on many factors: storage conditions, temperature, storage duration, etc. Thereby, the efficiency of pyrotechnic generators depends on observance of technological conditions of their manufacture as well as on the period and conditions of storage. Herein, a tendency of efficiency decreasing, sometimes by orders of magnitude, can be observed [4].

In this regard, laboratory technologies aimed at studying the yield of generators are particularly relevant. To implement an ideal case in which a technique for determining the efficiency of means of crystallization most fully takes into account different situations of impact on supercooled clouds and various characteristics of generators of ice-forming nuclei or crystals, we must provide the following conditions:

• The efficiency of a generator should be studied using direct simulation over the entire set of parameters, both parameters of the motion of the generator and the parameters of the environment in which the generator operates;

• Any dilution and transfer of selected aerosol samples must not be accompanied by changes in temperature and humidity;

• The nucleation and growth of ice crystals must occur under the direct simulation of the basic parameters of the seeding zone: temperature, humidity, and the spectrum of droplet size distribution.

The preference is given to aerodynamic tubes. Studies of generators in aerodynamic tubes make it possible to acquire reliable information on the efficiency of a particular generator in strictly controlled measurement conditions.

II. THE TECHNIQUE FOR THE DETERMINATION OF EFFICIENCY OF ICE-FORMING AEROSOL GENERATORS AND EXPERIMENTAL DATA

The aerodynamic stand for the testing of full-size generators of ice-forming aerosols is designed in the Institute of Electronic Engineering and Nanotechnologies of the Academy of Sciences of the RM with the participation of the Special Service for Active Influences on Hydrometeorological Processes of the RM.

This aerodynamic stand allows testing any type of pyrotechnical generators of ice-forming aerosols, which are used at present both in operations on protection of agricultural crops from hail damage and in operations (experiments) on modification of precipitation.

It should be noted that the simulation of conditions of the flight of a rocket using an aerodynamic stand is also caused by the fact that the ice-forming activity of aerosols is affected, to different extents, by many factors. One of them is the ratio of the velocity of the generator to the velocity of discharge of a gas-vapor stream from the nozzle of the generator. In addition, the yield of active ice-forming particles of AgI heavily and monotonically depends on this parameter.

The main difficulty in these experiments is to form particles, which are adequate to really used particles by their physicochemical and, accordingly, ice-forming characteristics. A practically significant parameter of artificial crystallization, which is necessary to characterize their performance under real conditions of impact, is the yield of ice-forming particles in a temperature range of $(-5 \div -15)^{o}C$.

It should be noted that, despite the considerable number of countries implementing projects on AIs, laboratories of this level are scarce in Europe (Russia, Bulgaria).

The aim of this work is to experimentally reveal the dependence of the main parameter of rocket generators, i.e., yield, on external factors, i.e., temperature, time, storage conditions, etc., under laboratory conditions with a maximal consistence of model conditions to real conditions of the flight of a rocket in a potentially hazardous cloud upon seeding with an ice-forming reagent.

To simulate real conditions of the operation of generators, the technique was based on the use of a stand prepared of a small horizontal aerodynamic tube (HAT) designed and constructed at the Institute of Electronic Engineering and Nanotechnologies, Academy of Science of Moldova.

The diameter of the HAT d = 330 mm; the length L = 9 m. In the Eiffel chamber (d = 500 mm, L = 3 m), a device for taking samples of ice-forming aerosol from the air flow is placed. The general arrangement of the aerodynamic tube in two project views is shown in Figs. 1 and 2. In the front part of the aerodynamic tube, in front of the Eiffel chamber, an access panel is arranged for the installation of full-size generators of ice-forming aerosols and large fragments of samples of pyrotechnical compositions with reagents. The flow velocity (m/s) in the HAT is determined by the method of measurement of gas dynamic pressure using a "Pitot tube."

Fig. 1. A fragment of the HAT.

Fig. 2. A fragment of the HAT with a system for sampling and dilution of aerosol.

To ensure a correct representativeness of the aerosol sample in the aerodynamic tube, we carried out special experiments to study the distribution of air velocity in the tube. Figure 3 shows the results of the air velocity distribution over cross section in the zone of the aerosol generator at a flow rate in the working part of 30 m/s.

It is evident that, for the used installation, the axis of symmetry in the horizontal direction almost coincides with the cross-section center.

For better homogenization and mixing of the aerosol and the air, at a certain distance behind the generator, we installed a special unit, i.e., turbulator intended for the intensive stirring of the aerosol plume in order to obtain a uniform aerosol concentration over the cross section at the sampling point. The estimation of the uniformity of the distribution of aerosol concentration over the cross section showed that the ratio of the concentration at the center to the concentration at any point of the cross section varies within 10%.

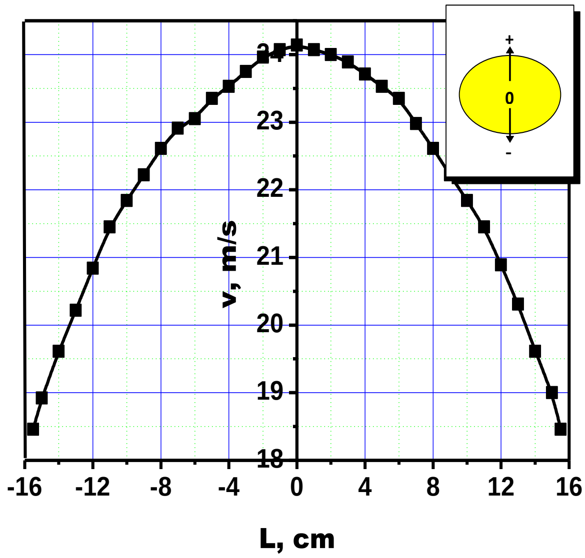

Fig. 3. Distribution of air velocity in the cross section of the aerodynamic tube

The system of sampling and dilution allows representative sampling of the aerosol generated by a generator in the air flow of the HAT. The intake is placed in the Eiffel chamber. The intake consists of a stainless pipe exposed at the center of the tube with holes directed toward the flow, a piping system, and a syringe. Since the difference between atmospheric pressure and dynamic pressure is usually no more than 1%, the transfer of the sample from the tract of the aerodynamic tube into a mixing chamber is carried out isothermally and with a constant humidity.

To prevent the suppression of the activity of ice-forming particles due to the effect of "re-seeding", it is necessary to obtain an optimum concentration of crystals in the mixing chamber, which would provide a statistically significant result of the experiment. In order to reduce the aerosol concentration, the sample had previously been dissolved in a cube with a volume of 1000 and 125 l, respectively (Figs. 1, 2).

The nucleation and growth of ice crystals occurs in a supercooled fog produced in the working volume of the mixing chamber. In the capacity of a mixing chamber in the stand under discussion, we used an ILKA KTLK-1250 climate chamber with a working volume of 1200 l manufactured in the German Democratic Republic. In accordance with the objectives of the experiments, the camera was modified as follows.

- Access holes were made in the door for the placing and removal of microthermostats.
- A system for introducing a measured sample of the active aerosol into the working volume was prepared.
- A fan to mix the air to reduce temperature gradients was installed in the working volume.
- A temperature and humidity sensor was installed.
- In the upper part of the cloud chamber, a lighting unit was installed; it generated a beam of light for the visual observation of the process of formation of fog, variation in its density, and formation of ice crystals.

Supercooled fog in the chamber is created by the injection of hot vapor, which condenses to form water aerosol with a modal diameter of droplets of about 4 μm.

Given the linear dimensions of the chamber, the vertical temperature gradient in the chamber does not exceed 0.02 deg/cm; the horizontal gradient, 0.005 deg/cm.

The initial water content of fog depends on the duration of the introduction of vapor; it was 0.4-3.0 g/m^3.

The accuracy of temperature measurement in the chamber working volume is ±0.1^{o}C.

For the experiment (measurement) temperature, we take the temperature settled in the chamber working volume after the formation of fog in it, before the introduction of the aerosol sample.

The lifetime of vapor fog in the chamber for an initial water content of 1÷2 g/m^3 is 2÷3 min.

Fog in the cloud chamber is generated by the condensation of a hot water vapor being introduced into a cooled volume.

The activation of samples of ice-forming aerosol is carried out in the chamber at specified temperature levels up to T = -20^{o}C.

Ice crystals that are formed on introduced nuclei grow to sedimentation sizes and are recorded at the bottom of the chamber using a thermostat. The quantity of the microthermostats is determined by objectives of the specific experiment. According to the number of crystals formed, knowing the characteristics of the equipment and consumption data for the generator, we can calculate the yield of nuclei. It should be noted that the time of the manifestation of ice-forming nuclei depends on temperature; in addition, the kinetic constant decreases with decreasing temperature, and the time of the manifestation of a given fraction of nuclei increases and is about 3 min at -5^{o}C.

The duration of one measurement (experiment) using the aerodynamic stand, which consists in the measurement of the yield of active ice-forming particles, is 30-40 min at a given temperature.

For the determination of the yield of active ice-forming particles according to this technique, it is necessary to take into account a number of factors, the disregard of which can significantly distort results:

- presence of significant temperature gradients in the chamber working volume;
- inhomogeneity of water content of supercooled fog;
- run-to-run reproducibility of fog parameters;
- local supersaturation of water vapor upon the introduction of aerosol in the chamber;
- coagulation of aerosol particles in the process of formation and introduction into the chamber, their precipitation on chamber walls, injector, and feeding hoses.

The estimation of accidental errors of measurements showed that the error of a single measurement is ±15% in a temperature range of -10 to 20^{o}C and ±30% at the fog temperature of -5^{o}C. As the temperature decreases, the error is reduced. The total systematic error is ± 3% and can be ignored in the calculations.

Based on the designed aerodynamic stand and developed procedure of testing of ice-forming pyrotechnic compositions, we carried out experiments on the determination of the practical yield of active particles of a pyrotechnic composition for different series of antihail rockets, which were produced in different years and used for active impacts on hail-forming processes by the Special Service for Active Influences on Hydrometeorological Processes of the Republic of Moldova in the respective years (Fig. 4).

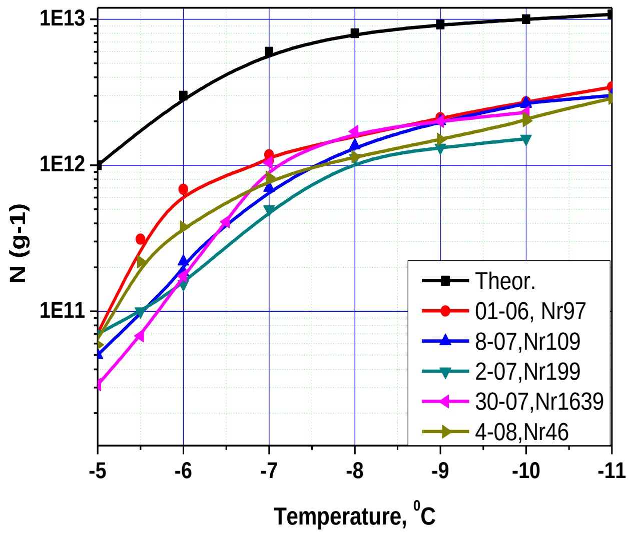

Fig. 4. Temperature dependence of the yield of active particles of a pyrotechnic composition of antihail rockets.

Analysis of Fig. 4 shows that, in general, the behavior of temperature dependence remains the same. However, depending on the year of manufacture of the rocket (the rocket's storage time at deposit), an interesting relationship is observed: the "older" the pyrotechnic composition, the lower its yield. On the basis of general considerations, this dependence of the generator yield is clear. Under the influence of external factors (temperature, time, storage conditions, etc.), irreversible changes take place in a pyrotechnic composition and lead to the experimentally observed decrease in yield of the generator of antihail rockets.

III. CONCLUSION

Based on the technique and devices that were developed and introduced into practice for studying the efficiency of crystallization means on the basis of silver iodide AgI, we analyzed the yield of really used generators of rockets of the "Alazan" type as a functions of external factors. This technique makes it possible to perform a direct simulation of the formation of aerosols and their interaction with the supercooled cloud environment, which gives the possibility to obtain adequate information about the efficiency of the generator in a real process of exposure. The technique is based on a direct aerodynamic modeling of the motion of a real generator in the air in an aerodynamic tube. It is shown that the efficiency of the pyrotechnic compositions based on silver iodide (AgI) decreases depending on storage time. The decrease in yield for given standard storage conditions is on the order of 10% per year.

The technique is protected by an author's certificate. To date, it has been used for the practical testing of generators of ice-forming aerosols based on Alazan' and Loza rockets used in our country. The developed technique and devices make it possible to systematically study the yield of mass-produced generators of rockets that are used in the practice of active influences (Alazan', Loza).

Thus, the totality of the studies is a solution to the important scientific and technical problem, i.e., the rapid determination of the yield of real generators used in Moldova in the practice of active influences on supercooled cloud environments to prevent hail. The results of these studies based on the experimentally measured value of yield give the possibility to correctly calculate the minimum number of rockets required for the processing of a potentially hail-hazardous cloud.

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