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SECOND EXPERIMENT ON THE INVESTIGATION OF ION-MOLECULAR REACTION CONSTANTS DIRECTLY IN THE IONOSPHERE

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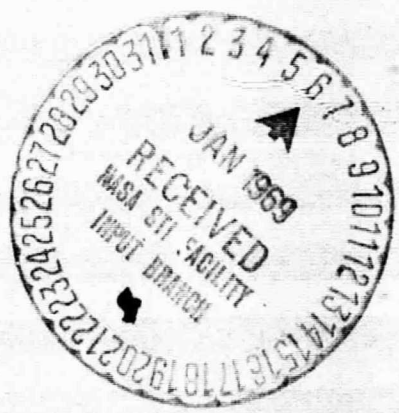

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SUMMARY

This note describes a new and improved experiment, involving besides the basic ion-molecular reactions, a new set of reactions which participate in the formation of NO^+ and O_2^+ ions at the expense of neutral molecules O_2 and N_2. Good agreement is shown with Fehsenfeld data relative to the ratio γ_2/γ_1, thus far considered to be the most reliable. The apparatus installed aboard the geophysical rocket performing the experiment is described.

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* *

The ion-molecular reactions play an important part in the ionization-recombination cycle of processes in the Earth's ionosphere. It was shown in [1], that laboratory measurements indicate a significant scattering in the obtained values of constants γ of the basic ion-molecular reactions:

$$O^+ + N_2 \rightarrow NO^+ + N, \quad (1)$$

$$O^+ + O_2 \rightarrow O_2^+ + O. \quad (2)$$

This scattering is of one order or more. As a consequence of that, it is not clear, which values γ_1 and γ_2 must be used for the ionosphere investigations.

A method of investigation of constants of the two fundamental ion-molecular reactions (1) and (2) was briefly described in the work [2] by way of release of fixed portions of gas in the immediate vicinity of the radio-frequency mass-spectrometer's analyzer during the flight of the rocket; presented in the same work are also the results of the first experiment of such kind,

conducted in 1962 with an AR (air release) device at the altitude of about 400 km. Following was the estimate obtained for the constant ratio of reactions (1) and (2):

$$\gamma_2/\gamma_1 \geqslant 10. \qquad (3)$$

In March 1967 at middle latitudes of the European USSR a launching was conducted at noontime of a geophysical rocket equipped with the following devices: an MX6407M radio-frequency mass-spectrometer for the investigation of the ionic composition of the atmosphere and an AR-device marked AR-2. The latter consisted of a glass container with air of about 500 cm^3 capacity of the opening and ejecting installations. A block-diagram of mutual disposition of the container and mass-spectrometer is shown in Figure 1. The container was provided with a capillary tube, which opened up at a preassigned altitude at a specific moment of time. The gas outflow through the capillary lasted 7 seconds, after which the container was ejected with the aid of a special device, so as to preserve the vacuum cleanliness of subsequent mass-spectrometric measurements. Prior to capillary "spout" opening, the air in the container was at atmospheric pressure, the diameter of the capillary being 0.7 mm.

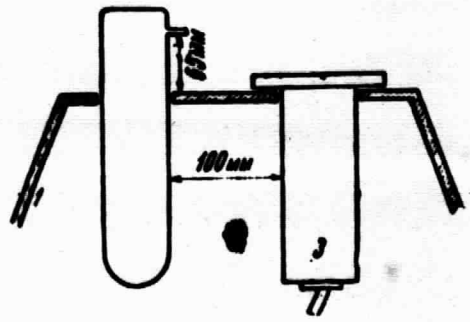

Fig.1. Sketch showing the respective position of the AR-container and mass-spectrometer during the experiment. 1) upper part of instrument compartment; 2) Glass container with air; 3) Analyzer of the MX6407M m.s.

In the described experiment air release was performed near the summit of rocket trajectory, at about 170 km altitude. A sharp variation was registered in the character of the spectrum of ions, recorded by the mass-spectrometer, which corroborates the wear and tear of the AR-device. The beginning of air release (spout opening) and its end (container ejection) were controlled by a special telemetry channel.

As was shown by analysis of the obtained spectra, during the first seconds following the beginning of air release, the gas cloud was so dense that it practically shielded off the mass-spectrometer from the surrounding medium; this is why peaks of atmospheric ions/, as well as of ions, conditioned by neutral particles of the released gas, were absent in the first spectrum. For analysis

we used the second spectrum, where shielding by cloud particles was already substantially weaker, sufficiently good peaks being already observed. The estimate of the magnitude of the shielding was conducted by the ion O^+ peak. Inasmuch as the released gas did not contain particles of atomic oxygen, it was assumed that the peak of mass 16 on spectrum No.2 was due to concentration of atmospheric ions O^+, diminished on account of the shielding effect. Comparison of intensity $I^{(2)}(O^+)$ in spectrum No.2 with the quantity $I_{atm}(O^+)$ registered by the mass-spectrometer for a few seconds prior to operation of the AR-device has shown that the shielding factor K constituted 1.76. In correspondence with this, the peak intensities of ions NO^+ and O_2^+, registered on the spectrum prior to release, were decreased by a factor of 1.76. The values of $I_{atm}^{(2)}(NO^+)$ and $I_{atm}^{(2)}(O_2)$, thus obtained, were interpreted as the contribution to peak intensities of masses 30 and 32 in spectrum No.2 at the expense of atmospheric ions NO^+ and O_2^+ taking into account the effect of the shielding. The difference between the really registered intensity of these peaks in spectrum No.2, $I^{(2)}(NO^+)$ and $I^{(2)}(O_2^+)$ and the values of $I_{atm}^{(2)}(NO^+)$ and $I_{atm}^{(2)}(O_2^+)$, was interpreted as the result of ion-molecular reactions between molecules N_2 and O_2 of the released air and the atmospheric oxygen ions:

$$I_{AR}^{(2)}(NO^+) = I^{(2)}(NO^+) - I_{atm}^{(2)}(NO^+); \quad (4)$$

$$I_{AR}^{(2)}(O_2^+) = I^{(2)}(O_2^+) - I_{atm}^{(2)}(O_2^+). \quad (5)$$

Inasmuch as the lifetime of particles in the protracting field of mass-spectrometer's analyzer is short, just as was the case in the work [2], it was assumed that peak intensities of ions NO^+ and O_2^+, due to the operation of the AR-device, are proportional to the rate of these ions' formation as a result of ion-molecular reactions. When accounting for only two basic reactions (1) and (2), this leads to the following expression for the ratio of their constants γ_1 and γ_2:

$$\gamma_2/\gamma_1 = I_{AR}(O_2^+)[N_2]_{air}[O^+]_{atm}/I_{AR}(NO^+)[O_2]_{air}[O^+]_{atm}, \quad (6)$$

where $[N_2]_{air}$ / $[O_2]_{air}$ is the ratio of nitrogen and oxygen molecules' concentrations in the released air, equal to 3.7, and $[O^+]_{atm}$ is the concentration of atomic oxygen ions in the surrounding atmosphere. The value of the peaks'

ratio $I_{AR}^{N_2}(O_2^+)/I_{Ak}^{N_2}(NO^+)$ in the given experiment after having accounted for the contribution of atmospheric ions and the influence of the shielding effect, was found to be 2.7, making the constant ratio γ_2/γ_1 in (6) equal to ten ($\gamma_2/\gamma_1 = 10$).

However, at the present time the laboratory data on constants of ion-molecular reactions lead to the conclusion that, besides processes (1) and (2), other reactions can also participate in the formation of ions NO^+ and O_2^+ from neutral molecules N_2 and O_2, which are the following:

$$N_2^+ + O_2 \rightarrow NO^+ + NO; \quad (7)$$
$$O_2^+ + N_2 \rightarrow NO^+ + NO; \quad (8)$$
$$N^+ + O_2 \rightarrow NO^+ + O; \quad (9)$$
$$N_2^+ + O_2 \rightarrow O_2^+ + N_2; \quad (10)$$
$$N^+ + O_2 \rightarrow O_2^+ + N. \quad (11)$$

Reactions (7) and (8) are little effective (γ_7 and $\gamma_8 \leqslant 10^{-13}$ cm$^3\cdot$sec^{-1} [3] and cannot compete with reaction (1). Estimates based upon the relative concentrations of ions $[N_2^+]$ / $[O^+]$ and $[N^+]$ / $[O^+]$, registered in the spectrum prior to the release, show that for $\gamma \approx 10^{-10}$ cm$^3\cdot$sec^{-1}, reaction (10) does not contribute substantially to the formation of ions N_2^+ in the conditions of the experiment, while the contribution of reactions (9) and (11), respectively to the formation of NO^+ and O_2^+, is substantial and must be taken into account. For constant values $\gamma_9 \approx \gamma_{11} \approx 5\cdot10^{-10}$ cm$^3\cdot$sec^{-1}, taken in accordance with laboratory measurements of [4], such an accounting was actually made and resulted in a refining of the obtained constant ratio: $\gamma_2/\gamma_1 = 1.3\cdot10$. The accounting of possible errors of measurements allows us to consider that the results of the experiment yield the following value range for constant ratio sought for:

$$\gamma_2/\gamma_1 = (1.0 - 1.5)\cdot10, \quad (12)$$

The obtained value is related to the altitude of about 170 km, which is characterized in daytime unperturbed conditions by a temperature of $\approx 1000°$K, in accordance with [5]. The found value of γ_2/γ_1 agrees well with the results of the first experiment with the AR-device [2], with the then obtained result $\gamma_2/\gamma_1 = 10$. Analysis of ionospheric estimates, mainly referring to high altitudes in the ionosphere (F_2-region) has shown [1] that there the constant

ratio of reactions (1) and (2) constitutes about 10 with a precision to a factor of 1.5 to 2. Comparison of the results of laboratory experiments by Fehsenfeld [6, 7], which now appear to be the most reliable, leads to ratio $\gamma_2/\gamma_1 = 1.3 \cdot 10$ at a temperature of the order of 300°K. Comparing these values among themselves one must, however, be cautious, inasmuch as the question of temperature dependence of constants of ion-molecular reactions so far has not been solved.

It should be noted that despite the good agreement of the obtained ratio γ_2/γ_1 with data by Fehsenfeld et al, the given experiment does not relieve us from the question of applicability of laboratory values of ion-molecular reactions' constants to ionosphere investigations. If there is in the upper atmosphere a substantial quantity of excited molecules O_2 and N_2, participating in the discussed reactions, the real rate of these reactions may substantially depart from those obtained in laboratory. Discussed in the described experiment is also the interaction rate of O^+ with unexcited molecules of the gas, inasmuch as the lifetime of these molecules after their emergence from the container is very short. However, the participation in the investigated reactions of real atmospheric ions with an ion temperature corresponding to the given altitude, renders the experiment conditions closer to the real ones in the ionosphere. This allows us to obtain during the performing of several gas releases at various altitudes (with different T_i) a dependence of the investigated constants on the ionic temperature. Besides, measurements at altitudes with different ratio $[N^+]/[O^+]$ allows us to find all the constants of interest to us without assigning ourselves the laboratory values of quantities γ_9 and γ_{11}. Although the reproduction of laboratory experiments is incomparably higher than similar isolated experiments, it appears that the latter may be useful for the understanding the extent, to which laboratory measurements of the values of γ are applicable to ionosphere investigations, and also for filling certain still present gaps in laboratory data.

*** THE END ***

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