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NATURAL IONIZATION IN GASES

By James W. Broxon

Abstract

Measurements of the *pressure variation* of the natural ionization in air, nitrogen, oxygen and carbon dioxide contained in a steel sphere of one foot inside diameter, made by H. F. Fruth for pressures from 1 to 75 atmospheres, were repeated with slight modifications. Results obtained corresponded with those of Fruth with the following exceptions: (1) although the pressure rate of variation of the natural ionization at high pressures was very small (0.4 to 1.0 ion/cc sec. atm.) no abrupt change in the rate of variation and no true "saturation" value of the ionization were observed; (2) no "anomalous" behavior was noted in the case of commercial nitrogen. Measurements to detect the *variation with size of chamber* were carried out with spherical iron ionization chambers of 7, 9, and 12 in. respective internal diameters. The rate of production of ions per cc at high pressures did not vary unidirectionally with the size of the ionization chambers, but was least in the 9 in. sphere. To study the *effect of the lining* the experiments were repeated with oxygen in each of the spheres lined with aluminum of 1/16 in. thickness, and in the 12 in. sphere lined with copper of 1/16 in. thickness. The aluminum lining increased the ionization at low pressures, and decreased its pressure rate of variation at high pressures. The copper lining reduced the absolute value of the ionization at high pressures. The *effect of water vapor* on the natural ionization in aged, dust-free air, or upon its variation with pressure was found to be negligible. Both the absolute and the fractional *variation of the natural ionization with time* were found to depend upon the pressure, the average value of the former during a 24 hr. period being about twice and that of the latter being about 1/5 as great in oxygen at 55.4 atm. pressure as in air at atmospheric pressure. Very slight evidence of a singly periodic diurnal variation in air at atmospheric pressure was observed. Attempts to determine the *effects of slow chemical reactions* upon the natural ionization in air and oxygen at atmospheric pressure led to negative results.

Introduction

THIS paper has to do with the ionization occurring in closed vessels apart from the action of definitely recognized and localized ionizing agents, a problem first attacked from the present point of view by C. T. R. Wilson[1] and Elster and Geitel.[2] In their work on the variation of the natural ionization with pressure, it was found by Downey[3] in the case of air, and by Fruth[4] in the cases of "purified" air and oxygen,

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[1] C. T. R. Wilson, Proc. Camb. Phil. Soc. **11**, 32 (1900); Roy. Soc. Proc. **A68, 151** (1901).

[2] H. Geitel, Phys. Zeits. **2**, 116 (1900);
J. E. Elster and H. Geitel, Phys. Zeits. **2**, 560 (1901).

[3] K. M. Downey, Phys. Rev. **20**, 186 (1922).

[4] H. F. Fruth, Phys. Rev. **22**, 109 (1923).

that the natural ionization in a gas in a spherical steel container increased with the pressure until pressures of about 46 and 52 atmospheres, respectively, were attained, and remained constant as the pressures were still further increased. Fruth found a similar saturation value in the case of carbon dioxide at 61 atmospheres. His results show no such saturation value in the case of nitrogen either as prepared commercially or in the laboratory. Moreover, no saturation value was observed in the case of oxygen and nitrogen which had been mixed in approximately the same ratio as they occur in air.

It was with the particular object of further investigating the occurrence of these saturation values and of discovering the reason for the "anomalous" behavior of nitrogen, that the present work was undertaken at the suggestion of Professor W. F. G. Swann.

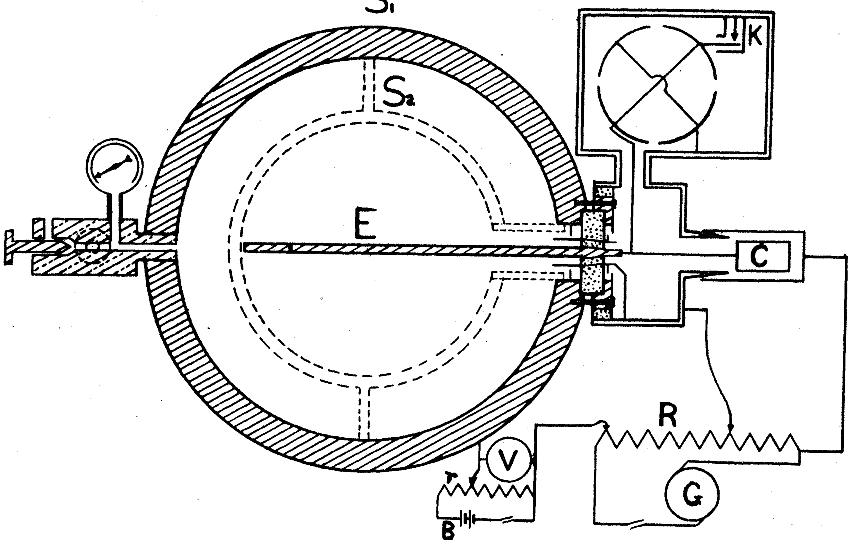

FIG. 1. Diagram of apparatus.

APPARATUS AND MANIPULATION

The ionization chamber consisted of the same iron sphere of one foot internal diameter, with walls of one inch thickness, which was used by Fruth, and the experimental arrangement corresponded closely with that described by him, certain modifications being made as shown in Fig. 1. The guard system was slightly modified, lead was substituted for the red rubber gasket, and the compensating condenser, C, was fixed. Observations and calculations were also made in essentially the same manner as in Fruth's work, all the precautions listed by him being taken in this instance.

In addition to the original sphere of 12 in. internal diameter, two iron spheres were cast, their thickness being 1/4 in. and their diameters 7 in. and 9 in., respectively. These could be inserted individually within the 12 in. sphere and held concentric with it by means of collars and a connecting pipe, as shown by the dotted S_2 in Fig. 1, E being made correspondingly shorter. As in the case of S_1, these spheres were actually cast as hemispheres and bolted together, but no attempt was made to make the inner spheres gas-tight, the pressure inside them being everywhere the same as that inside S_1.

In the present case, electrometer fibers from 2 to 5 times as sensitive as those used by Fruth were employed; three individual sets of readings were taken in practically every instance at each pressure; and the gases were passed first through the drying tube and then through the dust remover. The calibration constant, K, for a given arrangement of the ionization chamber, was usually considered at all pressures[5] to have the same value as at atmospheric pressure. In the case of carbon dioxide, however, a considerable variation was observed, and K was determined at each pressure. Saturation currents were assured by using ionization chamber potentials such that an increase of less than 0.3 percent in the currents resulted when the potentials were doubled.

No elaborate system of shielding was adopted. The thick walls of the ionization chambers themselves provided a certain amount of shielding from external radiations. In one instance, with the 9 in. sphere in position and filled with oxygen at a pressure of 200 lbs. per sq. in., measurements of the natural ionization were made before and after the placing of boxes of lead storage battery plates in the vicinity of the ionization chamber. These plates provided roughly a 6 in. lead wall subtending a solid angle of approximately $4\pi/3$ at the center of the chamber. With due consideration for its variation with time, the natural ionization could not be considered to have been decreased by more than $3\frac{1}{2}$ percent by the lead shield. An actual decrease, however, appears to have occurred.

The hydrogen and hydrogen sulphide used were formed in Kipp generators by the action of hydrochloric acid upon zinc and upon ferrous sulphide, respectively. The other gases used were purchased from commercial producing companies, with the exception of air which was compressed in the laboratory.

[5] It should be noted at this point that throughout this paper all pressures referred to in terms of lbs per sq. in. are pressures *in excess* of atmospheric pressure.

RESULTS AND DISCUSSION

Pressure Variation. In order to determine the variation with pressure of the natural ionization in certain gases, the natural ionization occurring in air, oxygen, nitrogen and carbon dioxide was measured with the 12 in. sphere as ionization chamber, at various pressure intervals between atmospheric pressure and pressures in the neighborhood of 1000 lbs. per sq. in. The results are shown in Figs. 2, 3 and 5.

In view of the announced purpose of this research, perhaps the most noticeable feature of the results obtained is the fact that in no instance was a true saturation value observed. According to Fruth's[4] results a saturation value of the ionization should have been obtained at 53 atmospheres or about 779 lbs. per sq. in., in the cases of oxygen and of air contained in the 12 in. sphere. However, although the above pressure was exceeded by from 13.6 to 22.5 atmospheres with both oxygen and air in all the cases in which the pressure variation with these two gases was investigated, the curves in no instance became horizontal nor was there evidence of a sharp break at any point. On the other hand, all the curves obtained are smooth and are everywhere concave downward, i.e., toward the pressure-axis. It is seen, therefore, that with both air and oxygen the slopes of the P-n curves decreased gradually with pressure, so that the curves may be considered to be approaching saturation values. However, the pressure rates of change of slope at the highest pressures are very small and it is only possible to conjecture as to whether the curves would finally reach saturation values, whether their slopes would continually decrease without vanishing, or whether the curves would become straight lines with constant small slopes. The gradual decrease in the curvature of these curves together with the form of the curves for nitrogen and carbon dioxide, tend rather to persuade one toward the latter view. The pressure curves obtained with nitrogen and carbon dioxide are practically straight lines at pressures above 800 and 500 lbs. per sq. in., respectively.

In their most important aspects, the results described in this paper agree with those of Downey and Fruth. In each case investigated, with whatever gas, size of ionization chamber or nature of lining, at the highest pressures obtained the rate of increase in the natural ionization with pressure was very small, being in no case greater than one ion (of one sign) per cc per sec. per atm. The values given in Table I for the least slopes agree very well with the values of Δq given in Table II of Dr. Downey's 1922 paper for the CD portion of her curves, as well as with the final slopes of Fruth's curves apart from the horizontal portions of his curves for air and oxygen. As nearly as can be determined, if we neglect these portions of Fruth's curves, we have as the ratios of the final slopes of his curves to the final slopes of the curves in this paper for the same gases in the unlined 12 in. sphere: 1.67 for air; 1.37 for oxygen; 1.52 for nitrogen; 1.67 for carbon dioxide. If his "saturation" values be neglected, then, it would appear that the final slopes of his curves are from 1 1/3 to 1 2/3 times as large as those of curves obtained similarly in this investigation. In other words, there is very close agreement.

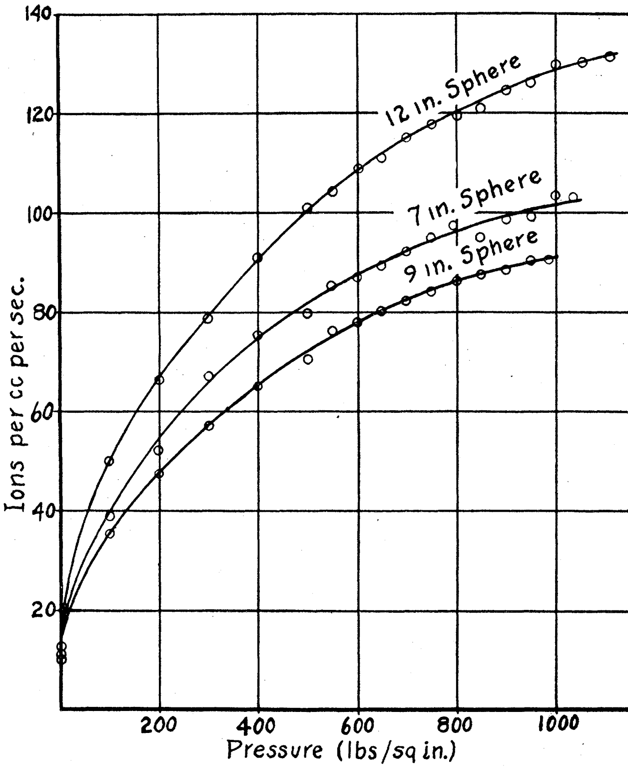

FIG. 2. Variation with pressure of the natural ionization in air.

It may be noted that some of the curves obtained by the writer are of the same general type as obtained by Wilson[6] for pressures up to 40 atmospheres. Evidence recently provided by Millikan[7] for the existence of a very penetrating cosmic radiation points toward the

[6] W. Wilson, Phil. Mag. **17**, 216 (1909).

[7] R. A. Millikan, Science **62**, 445 (1925); Nature **116**, 823 (1925); Proc. Nat. Acad. Sci. **12**, 48 (1926) especially p. 51.

probability that ionization due to this source at sea level should be very small, and of about the order herein obtained per atmosphere increase of pressure at the highest pressures. Swann[8] has drawn attention to the smallness of the effect to be expected from this source.

VARIATION WITH SIZE OF IONIZATION CHAMBER

The magnitude of the natural ionization occurring in a gas at high pressures and also its rate of variation with pressure, were found to depend upon the size of the ionization chamber. This dependence is readily seen upon inspection of Figs. 2, 3 and 4. One outstanding fact which was not expected is that at the higher pressures the curves for the 7 in. sphere lie between those for the 12 in. and 9 in. spheres. Thus the ionization (ions/cc sec.) must pass through a minimum as the size of the chamber is varied. This peculiarity exists at all pressures above 1 atm. excepting in the case of oxygen in the unlined spheres, in which

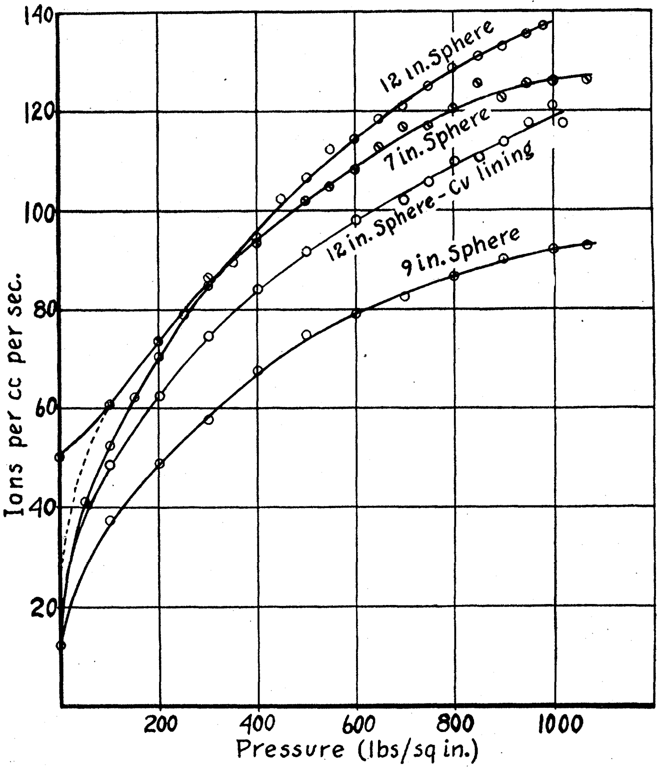

FIG. 3. Variation with pressure of the natural ionization in oxygen.

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[8] W. F. G. Swann, Nat. Res. Coun. Bull. **17**, 3, 65 (1922).

case the curve for the 7 in. sphere crosses that for the 12 in. sphere and the former has an abnormally high value at atmospheric pressure. If the ionization were chiefly due to soft radiations from the walls of the ionization chamber and if the magnitude of these radiations could be considered to be proportional to the area as long as the walls were made of the same material, then an increase in the number of ions per cc per sec. should be expected to accompany a decrease in the volume with its accompanying increase in the ratio of area to volume. A theoretical discussion of these matters will appear in a subsequent publication.

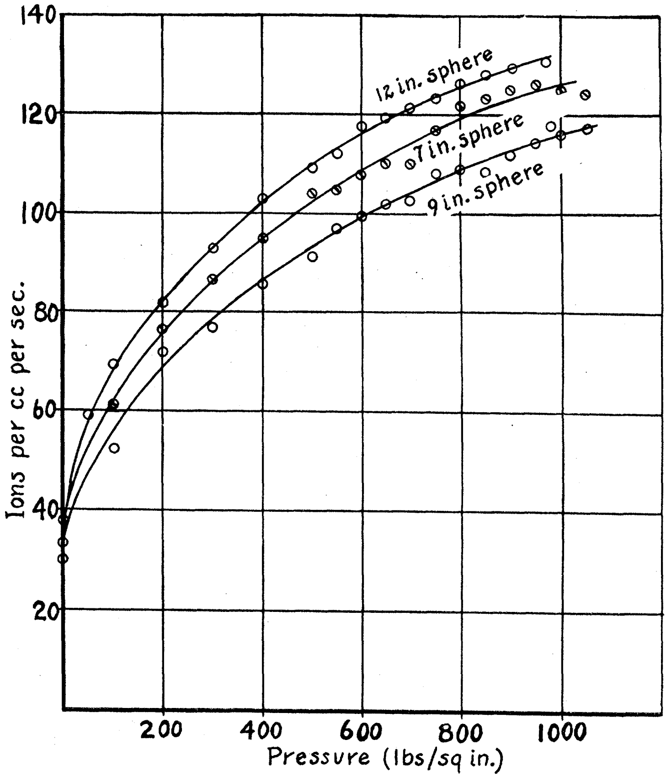

FIG. 4. Variation with pressure of the natural ionization in oxygen. Spheres lined with aluminum.

At atmospheric pressure, the rate of production of ions per cc in the 12 in. sphere was in each case slightly less than the corresponding ionization in the smaller spheres. However, without more extended series of readings with the various spheres at atmospheric pressure, this observation should not be considered to be well established.

One interesting observation is (see Table I) that the smallest slope or the least change in the number of ions per cc per sec. per atmosphere increase of pressure, apparently decreased slightly with the size of the spherical ionization chamber. The smallest value observed was about 0.18 ion/cc sec. atm. in oxygen contained in the 7 in. sphere at a pressure of 1070 lbs. per sq. in.

DEPENDENCE UPON KIND OF GAS

As has been mentioned previously, the curvature in the nitrogen and carbon dioxide curves appears to be quite definitely zero over considerable distances at their high-pressure ends. Although all the values at atmospheric pressure were of about the same magnitude, the values of the ionization in nitrogen and carbon dioxide at the higher pressures were considerably greater than the values at corresponding pressures in the other gases in the same 12 in. sphere.

It is seen upon reference to Table I that the least slopes of the oxygen and air curves are of about the same magnitude, whereas those for nitrogen and carbon dioxide are considerably greater and about equal. In fact, upon superposition of the nitrogen and carbon dioxide curves (Fig. 5), it is seen that they nearly coincide throughout, the latter being a little more concave at the lower pressures, while in the former the concavity is smaller but more persistent.

TABLE I

Least slopes of pressure-ionization curves.

Gas	Ionization Chamber	Lining	Least slope $\left(\frac{\text{ions}}{\text{cc sec. atm.}}\right)$
Air	12-in.	None	0.40
Air	9-in.	None	0.32
Air	7-in.	None	0.26
O_2	12-in.	None	0.65
O_2	9-in.	None	0.24
O_2	7-in.	None	0.18
O_2	12-in.	Al	0.50
O_2	9-in.	Al	0.41
O_2	7-in.	Al	0.40
O_2	12-in.	Cu	0.65
N_2	12-in.	None	0.97
CO_2	12-in.	None	1.00
Air + H_2O Vapor	12-in.	None	0.90

The curve for moist air shown in Fig. 5 was obtained by pouring 20 cc of distilled water into the 12 in. sphere, unlined, after which aged, dry, dust-free air was admitted to a pressure of 750 lbs per sq. in. Examination of the curve shows that there was no particular difference (so far as observations extended) between the ionization occurring in this case and that in dry air in the same chamber. Values at the higher pressures were slightly smaller in the former case than in the latter, but the divergence between the two curves is small and gradual. The final least slope of the air—water vapor curve is practically the same as that of the dry air curve at the same pressure. Therefore, water vapor evidently produces no decided effect upon the natural ionization in aged, dust-free air. It might, however, produce a considerable effect in the presence of dust particles.

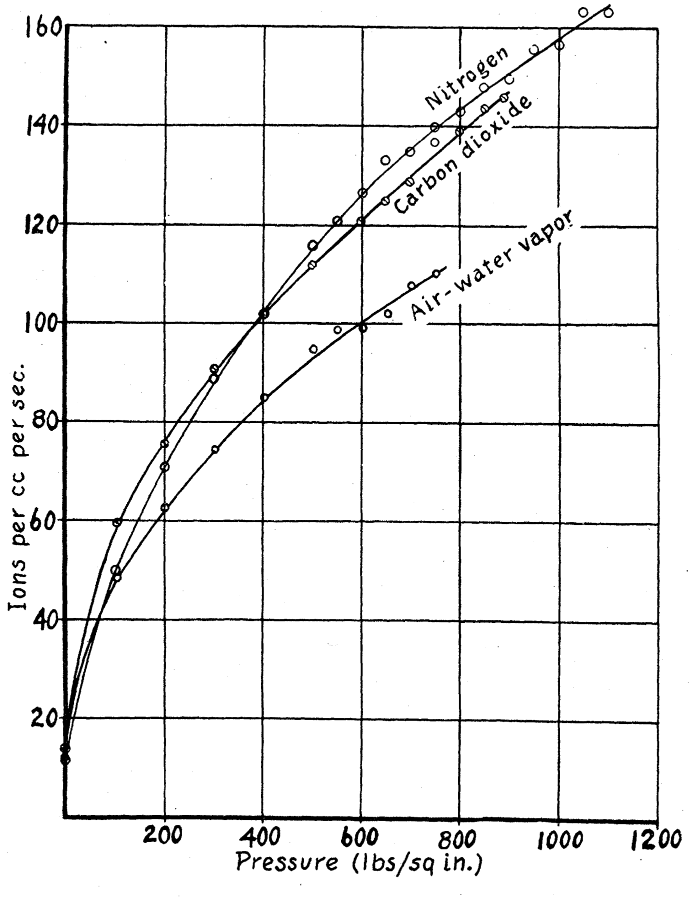

FIG. 5. Variation with pressure of the natural ionization in N_2, CO_2 and air saturated with water vapor.

Since no outstanding differences other than those mentioned above were found to exist among the values of the natural ionization or the P-n curves obtained for the various gases investigated, there evidently exists no special "anomalous" effect for nitrogen so far as this investigation is concerned. There remain, however, the larger slopes for nitrogen and carbon dioxide.

EFFECT OF LINING

The effects of the metals constituting the ionization chambers, upon the natural ionization in gases, particularly air, were early investigated by several experimenters,[9] especially by Campbell and McLennan. These investigations, however, were ordinarily carried out at atmospheric pressure. Evidence has been provided to show that the natural ionization does depend upon the material constituting the vessel, and also upon the particular sample of the material.

In Figs. 3 and 4 are the three P-n curves which were secured with oxygen in ionization chambers of practically the same size (12 in. diameter) but with different metals constituting their inner surfaces. It is seen that the two curves for the respective cases in which the iron sphere was unlined and in which it was lined with copper, start at practically the same point and have practically the same initial and the same final slope. The early intermediate curvature is rather greater in the case of the copper lined sphere, however, resulting in considerably lower values for the ionization at high pressures in this case.

With the chamber lined with aluminum, the value of the ionization at atmospheric pressure was much larger than in the other two cases. This particularly high value of the ionization at atmospheric pressure appears to have been characteristic of the aluminum lining inasmuch as it was observed with each of the three spheres (see Fig. 4). The curvature of the curve for aluminum, however, corresponds very closely with that of the curve for the copper lining for pressures below 600 lbs. per sq. in. and is more persistent than in the latter curve at greater pressures. The result, therefore, is that the aluminum curve has a smaller final least slope than do the other two curves for oxygen in the 12 in. sphere, and the final absolute value of the ionization is larger than with the copper lining.

As is shown by Fig. 4, the curves obtained with each sphere lined with aluminum are very regular and correspond closely. It might be concluded, therefore, that the aluminum gave rise to a rather bounteous supply of comparatively easily absorbable radiation. This suggests

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[9] A. Wood, Phil. Mag. **9,** 550 (1905);
N. R. Campbell, Phil. Mag. **11,** 206 (1906);
N. R. Campbell, Camb. Phil. Soc. Proc. **13,** 282 (1906);
J. C. McLennan, Phil. Mag. **14,** 760 (1907);
J. C. McLennan, Phys. Rev. **26,** 526 (1908).

an explanation in terms of the considerable secondary cathode radiation excited by gamma radiation in substances of light atomic weight.[10] The chief effect of the copper lining appears to have been the reduction of the absolute value of the ionization at high pressures. It should be noted that radiations from the iron which could have penetrated 1/16 in. of aluminum or copper would have affected the ionization occurring with these two linings.

Variation with Time

The variation with time of the natural ionization in the atmosphere is of interest in connection with problems concerned with the atmospheric potential gradient, the emanation content, the magnetic intensity, etc. Wood and Campbell,[11] McKeon,[12] Strong,[13] Wright,[14] Gockel,[15] Simpson,[16] Wulf,[17] Downey,[18] and others have investigated more or less exhaustively the variation with time of the natural ionization in air at atmospheric pressure. No general agreement appears to have resulted as to whether the diurnal variation is singly or doubly periodic or whether it exists at all.

In the present investigation of the variation at atmospheric pressure there was, as is designated by the dotted curves in Fig. 6, a decided similarity between fluctuations during the first two 24-hr. periods. The large general variations partook of a single periodicity with maxima in the early afternoon and minima in the early morning. The weather was turbulent or stormy during these periods. During the third period, when the sky was comparatively clear, the above periodicity did not appear. In view of previous work it is concluded that the present work, extending over a period of only 72 hrs., contributes very little toward the solution of the daily variation problem.

From the summary in Table II, it is seen that the average deviation from the mean at atmospheric pressure was of the order of $\frac{1}{2}$ ion/cc sec., or about 5 percent of the mean, whereas the largest variations were of the order of 2 ions/cc sec., or about 20 percent of the mean. Smaller fluctuations were observed by Swann[19] in the same laboratory at about the same time.

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[10] W. H. Bragg and J. P. V. Madsen, Phil. Mag. **15,** 663 (1908).

[11] A. Wood and N. R. Campbell, Phil. Mag. **13,** 265 (1907).

[12] T. F. McKeon, Phys. Rev. **25,** 399 (1907).

[13] W. W. Strong, Phys. Zeits. **9,** 117 (1908).

[14] C. S. Wright, Phil. Mag. **17,** 295 (1909).

[15] A. Gockel, Phys. Zeits. **10,** 845 (1909).

[16] G. C. Simpson and C. S. Wright, Roy. Soc. Proc. **A85,** 175 (1911).

[17] Benndorf, Dorno, Hess, v. Schweidler, Wulf, Phys. Zeits. **14,** 1141 (1913).

[18] K. M. Downey, Phys. Rev. **16,** 420 (1920).

[19] W. F. G. Swann, Jour. Franklin Inst. Oct. (1925).

Curve I of Fig. 6 shows that with oxygen at a pressure of 800 lbs. per sq. in. there was no periodicity corresponding to that at atmospheric pressure. The magnitude of the average deviation from the mean was about twice that in air at atmospheric pressure, and the magnitude of the maximum deviation was rather more than twice the largest deviation at atmospheric pressure. The average deviation from the mean at a pressure of 800 lbs. per sq. in. was only about 1 percent and the maximum deviation about 5 percent of the mean. The larger

TABLE II

Variations over 24-hr periods of the natural ionization at constant pressure.

Press.	No. of Readings	Mean no. of ions/ cc sec.	Ave. dev. from mean	Max. dev. from mean
800 lbs./in.^{2}	67	118.82	1.163	5.62
Atmos.	71	10.58	0.600	2.23
Atmos.	66	11.21	0.695	2.02
Atmos.	72	10.71	0.427	1.24

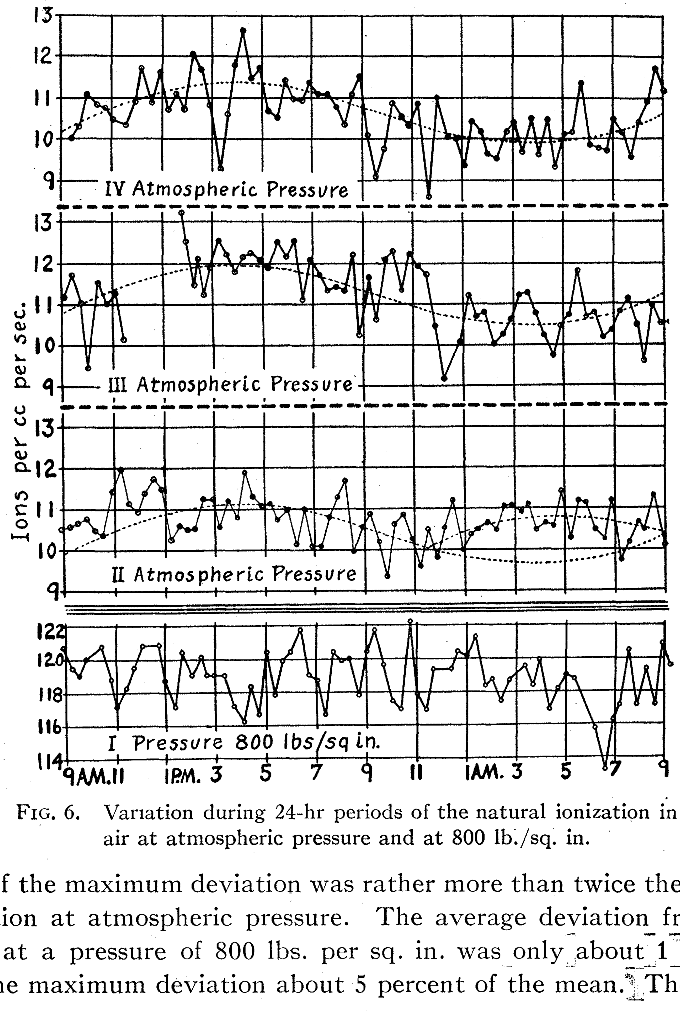

FIG. 6. Variation during 24-hr periods of the natural ionization in air at atmospheric pressure and at 800 lb./sq. in.

absolute values but the smaller fractional values of the variations occurring at the higher pressure may be explained in terms of fluctuations of soft radiations supposed to contribute to the total ionization.

INFLUENCE OF CHEMICAL ACTION

It has sometimes been suspected that chemical action might account for a part of the natural ionization in gases. With this possibility in mind, observations of the natural ionization occurring in air contained at atmospheric pressure in the 12 in. sphere were made before and after the admission of small quantities of hydrogen which would be expected to combine to a slight extent with the oxygen in the air. Also, observations of the natural ionization occurring within the 12 in. sphere in which had been placed pieces of silver foil and which had been filled with oxygen to atmospheric pressure were made before and after the admission of small quantities of hydrogen sulphide and also other quantities of silver.

In common with previous results,[20] those of the present investigation were negative so far as the detection of the dependence of the natural ionization in gases at atmospheric pressure upon slow chemical action is concerned. In certain cases there appeared to be slight increases in ionization at times coincident with increased chemical action. When the ionization values were plotted against the time, however, it was seen that there was practically no justification for the conclusion that the natural ionization was affected by the slow chemical action which resulted upon the admission of the different materials. That chemical action did occur upon admission of the hydrogen sulphide was evidenced by the tarnished condition of the silver and the brass parts of the apparatus subsequently observed.

In conclusion, the writer wishes to express his appreciation of the very general cooperation of the Physics faculty at Yale University where the experimental work was completed, and of the particularly kindly interest of Professor John Zeleny. Thanks are extended to Mr. C. J. Pietenpol who assisted in taking some of the readings, and to Professor W. B. Pietenpol for reading this paper. Finally, the writer desires to acknowledge his especial indebtedness to Professor W. F. G. Swann who suggested this research and who offered many helpful suggestions during the progress of the work.

HALE PHYSICAL LABORATORY,
UNIVERSITY OF COLORADO,
January 16, 1926.

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[20] K. H. Kingdon, Phil. Mag. **32**, 396 (1916).
N. R. Campbell, Phil. Mag. **9**, 545 (1905).