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Corresponding Member of the Academy of Sciences of the USSR S. E. FRISH and N. A. MATVEEVA

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Abstract

Full Text

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STUDY OF THE MECHANISM OF SEPARATION OF INERT GASES IN A DIRECT-CURRENT DISCHARGE

The passage of a direct electric current through a mixture of gases is known to cause their separation. There are two hypotheses concerning the mechanism of this separation: 1) the separation is caused by the transport motion of positive ions toward the cathode (⁀1); 2) the separation is caused by the transport motion toward the anode of neutral atoms arising under the influence of electron impacts (⁀2).

The existence of transport motion of ions in a discharge was directly confirmed by one of us together with Yu. M. Kagan (⁀3) through observation of the Doppler shift of ionic lines. Thus, the role of the transport motion of ions in the process of gas separation must undoubtedly exist. This point of view is further confirmed by the following fact: when gases are separated, near the cathode the concentration of the gas with the lower ionization potential increases. This is precisely what should occur in separation due to the transport motion of ions, since in the discharge the concentration of ions of the gas with the lower ionization potential is higher than that of ions with the higher ionization potential. Nevertheless, the existing experimental material is insufficient for a complete explanation of the mechanism of gas separation in an electric discharge. We therefore carried out a systematic investigation of gas separation in a discharge in mixtures of inert gases.

Fig. 1. Discharge tube

Most previously performed experimental work suffered from the significant drawback that the change in the composition of the mixture was determined from the change in spectral emission along the discharge column. Such a method, however, cannot yield quantitative information on changes in the concentration of the components, since the intensity of spectral lines is determined not only by the concentration of the given gas, but also by the excitation conditions, which,

Figure 2

Figure 2: Figure 2

when separation is present, are not the same in different parts of the positive column. To avoid this drawback, we withdrew gas samples from the near-electrode regions, which were then subjected to quantitative spectral analysis (ˆ4).

The discharge tube used in the work is shown in Fig. 1, where A and B are two vessels with capacities of 400 and 1600 cm^3, containing nickel electrodes; the vessels are connected by a tube 1 cm in diameter; the length of the tube was varied in different experiments from 25 to 150 cm; a and b are the sampling points. A series of probes was introduced into the tube for measuring the electrical characteristics of the discharge. The tube was supplied with direct current at a voltage of 1.5 kV. The direction of the discharge current was chosen so that in the smaller vessel A the concentration increa—

the concentration of the component that was present in the mixture in smaller quantities increased. All experiments were carried out with binary mixtures of helium, neon, and argon; as a rule, one of the components was taken in quantities considerably smaller than the other (as an impurity). The degree of separation was characterized either by the ratio of the impurity concentration in the enriched sample to its concentration in the initial mixture, C_{enr}/C, or by the ratio of the difference in impurity concentrations at the two ends of the tube to its concentration in the initial mixture, $\Delta C/C$.

Fig. 2. Dependence of the separation of an argon—helium mixture on the tube length.

The measurement error consisted of errors of spectral analysis and errors associated with the separation process itself. The arithmetic mean scatter of the values of the relative change in impurity concentration was 5-10% of the measured quantity.

When the discharge was switched on, the concentration of the components in vessels A and B at first began to change rapidly, and then reached a stationary value. This stationary value is evidently attained under the condition that the number of particles transported per unit time under the action of electric forces is equal to the number of particles diffusing in the opposite direction because of the concentration gradient. The time required to establish equilibrium increases linearly with increasing pressure of the mixture and depends little on the current strength and the concentration composition of the mixture. With increasing length of the discharge tube, the separation time increases. For a tube length of 150 cm, a pressure of 1.6 mm Hg, and a current strength of 50 mA, for an argon—helium mixture it reached 20 min. In addition to the time for establishing the stationary state, the mixing time of the separated mixture after switching off the discharge was also determined. It turned out that the mixing time is approximately equal to the separation time. For all subsequent

measurements, samples were taken from the discharge after the stationary state had been reached.

The following basic dependences were established:

1) The magnitude of the separation $\Delta C/C$ increases linearly with increasing tube length. Figure 2 gives the measurement results for argon–helium mixtures with an argon content of 1.3% for three tubes, in which the distance between the side arms for taking gas samples was 19, 34, and 61 cm; the pressure of the mixture was 1.5 mm.

2) With increasing pressure in the region of low pressures (0.5–1.5 mm), a noticeable increase in separation is observed. At higher pressures (1.5–4 mm), the degree of separation changes little. Probe measurements indicate approximately the same dependence of the longitudinal potential gradient on pressure.

3) With increasing discharge current, the degree of separation at first increases approximately linearly with the current, and then more slowly. At a sufficient current strength, a small impurity of the easily ionized component is practically completely separated from the principal component of the mixture. For example, in an argon–helium mixture, the argon content, initially equal to 3%, decreases at the anode, at a discharge current of 500 mA, to $\sim 10^{-3}$%. According to probe measurements, in a mixture of inert gases there is always a decrease in the longitudinal potential gradient with increasing discharge current.

4) With increasing concentration of the easily ionized impurity, the degree of separation decreases, and its dependence on pressure becomes less pronounced. Also, with increasing concentration of the easily ionized component, the degree of separation increases more slowly with increasing current strength. If the impurity is a difficult-to-ionize gas, separation occurs

to a very slight degree. It increases somewhat with increasing concentration of the difficult-to-excite component. For example, in the case of small admixtures of helium to argon, separation is practically absent, and only when the amount of helium reaches approximately one half is some separation observed.

5) The dependence of separation on the ionization potentials of the components of the mixture cannot be revealed in pure form, since when some gases are replaced by others, not only the ionization potentials but also the atomic weights change. However, in the region of not very large concentrations of the readily ionized admixture, the degree of separation for all the mixtures studied is the greater, the greater the difference between the ionization potentials of the components.

The results presented are qualitatively explained by the hypothesis that the main process causing gas separation is the transport motion of ions. The addition of a readily ionized component, even in small amounts, is known to lead to a

noticeable decrease in the electron temperature, which in turn leads to a strong decrease in the degree of ionization of the difficult-to-excite component. This explains the possibility of strong separation for small admixtures of the readily excited component. The greater the difference between the ionization potentials of the components of the mixture, the greater the difference in the concentration of their ions and the more strongly pronounced the separation effect.

For a more detailed explanation of the observed regularities, it is necessary simultaneously to take into account the dependence of the rate of transport motion of ions on the discharge conditions and the role of diffusion.

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CITED LITERATURE

1. M. J. Druyvesteyn, *Physica*, **2**, 255 (1935).

2. F. Skaupy, *Zs. f. Phys.*, **2**, 213 (1920).

3. S. E. Frisch, Yu. M. Kagan, *ZhETF*, **17**, 577 (1947); **18**, 519 (1948); *Izv. AN SSSR, ser. fiz.*, **12**, 358 (1948).

4. S. E. Frisch, E. Ya. Shreider, *Izv. AN SSSR, ser. fiz.*, **13**, 465 (1949); O. P. Bochkova, E. Ya. Shreider, *Spectral Analysis of Gas Mixtures*, 1955.

Note: Figure translations are in progress. See original paper for figures.