INVESTIGATION OF THE EQUILIBRIUM COMPOSITION OF A NITROGEN–HELIUM MIXTURE

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The results of experimental investigations of the equilibrium composition of a nitrogen–helium mixture are discussed.

When assessing the process of purification of helium from nitrogen, it is essential to have available reliable data about the equilibrium of the helium–nitrogen system.

In the Soviet and foreign literature, experimental data exist on the equilibrium of the nitrogen–helium system [1-5] but their use in engineering practice is made difficult by the fact that values of the equilibrium parameters differ significantly between themselves. Moreover, these data involve only the region of mixture states up to a temperature of 120°K. Nevertheless, when assessing a process for the separation of helium from a nitrogen–helium mixture it is necessary to deal also with temperatures in excess of 120°K and a wide range of change of concentrations.

In connection with this we were faced with the problem of establishing experimentally which of these data are the most reliable and also of extending the region of temperatures investigated, corresponding to their concentrations.

The experimental investigation of the process of purification of helium from nitrogen and the equilibrium composition of a nitrogen–helium mixture was carried out in a fractionating plant, shown schematically in Fig. 1.

The mixture of helium and nitrogen of given composition enters the membrane compressor MK-200 /2.5 from the gas holder with a capacity of 5 m^3, where it is compressed to the required pressure and is directed to the fractionating plant. The intertube space of the fractionating plant consists of two cavities – the working cavity 2 and the screening cavity 3.

Liquid nitrogen IV is fed into the intertube space of the gasifier, used as a reservoir, through the insulated pipeline. Its level in the cavity 2 is controlled by a hampsometer.

The stream of gaseous nitrogen V, formed by evaporation in cavity 2, is directed into the thermostat where it is heated to 293°K and it is discharged to the atmosphere through a GKF-6 type gas meter.

The screening cavity absorbs heat from the surrounding medium through the insulation 4 located in the metal jacket 5. The nitrogen, having evaporated in the cavity 3, forms a vapor VI which is also discharged to the atmosphere.

The fractionating plant 1 is made of pure copper tubing with a diameter of 10 × 1.0 mm and a working height of 100 mm.

The starting mixture of given composition enters either from below or above the fractionating tube 1.

The nitrogen from the mixture which is condensing on the inside surface accumulates in the condensate collector 6, which has a volume of $1.5 \cdot 10^{-4}$ m^3 and the concentrated gaseous helium is removed from the fractionating tube 1 from below or above. The products of separation of helium II and nitrogen condensate III are fed into a thermostat. The streams II and III, heated to a temperature of 293°K, are returned to the gasholder through the rotameter RS-3 and the GKF-6 meter. The total flow of II and III from a series

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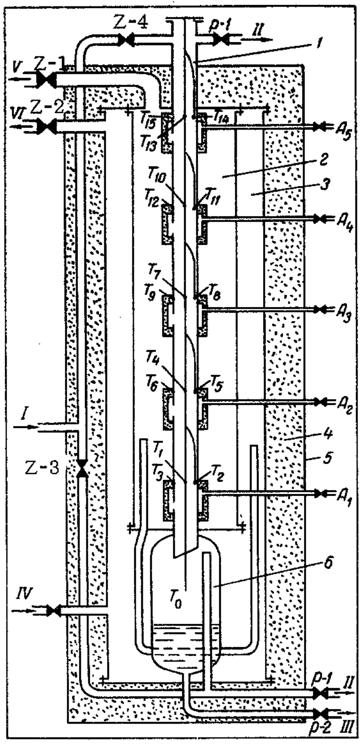

Fig. 1

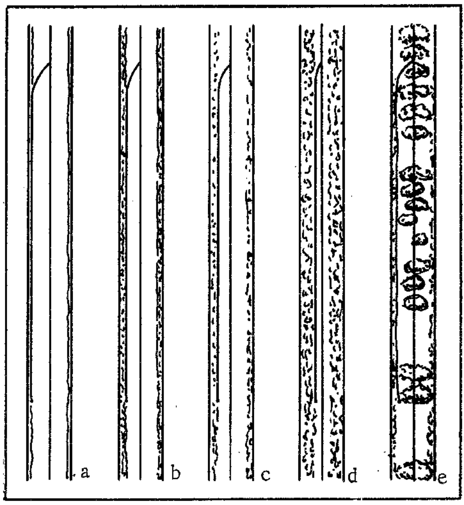

Fig. 2

Fig. 1. Layout of fractionating plant: A_1-A_5) analyzing valves; Z-1 –Z-4) shut-off valves; p-1 and p-2) throttle valves.

Fig. 2. Position of end of thermocouple in the case of straight-through flow (a, b, c and d) and counter-flow (e) of water and air.

of experiments is maintained approximately constant by means of rotameters and a bypass line mounted on the membrane compressor. The pressure in the tube and in the intertube space is measured by sampling manometers. Temperatures were measured by copper–constantan thermocouples, an R-306 potentiometer and an M 195/1 galvanometer.

The thermocouples were made from wire of diameter 0.05 mm.

Temperatures were measured in the main body of the vapor–gas flow at the surface of the condensate film facing the vapor–gas flow and at the wall; they were also measured at several points of the chamber wall. The temperature at the surface of the condensate film was measured by a thermocouple "floating" on its surface. The length of the free section of the thermocouple was established experimentally, contact being provided by the end of the thermocouple with the surface of the draining liquid, for which a slide-wire was stretched through the center of the drainage tube and a double slightly twisted wire made from the same material as the thermocouple was secured. Under the elastic force of the wire, the end of the thermocouple was pressed against the wall of the tube when no liquid was flowing. When liquid was flowing, which was fed in from above, the wire was deflected from the wall. Visual observations permitted it to be established that the nominal length of the free section of the wire must be not less than 120-150 mm. With smaller lengths and by the action of the elasticity of the wire, the end of the thermocouple remained pressed against the wall of the tube even with a relatively large flow of liquid.

Tests were conducted on a water–air system with both straight-through and counter-flow of the latter. The location of the end of the wire for different thicknesses of the draining film is shown in Fig. 2a, b, c and d. In these tests, the gas flow was from top to bottom, i.e. direct flow with the liquid.

In the case when the gas was moving from bottom to top, the tube started to function in the gas-lift cycle (Fig. 2e). Even in this cycle, it can be seen clearly that the end of the wire is located on the surface of the liquid facing the gas flow. The ratios found in the water–air system between the length of the free section of the thermocouple wire and the surface of the liquid film were later verified visually also in liquid nitrogen. Observations on the behavior of the thermocouple under these conditions showed that the ratios

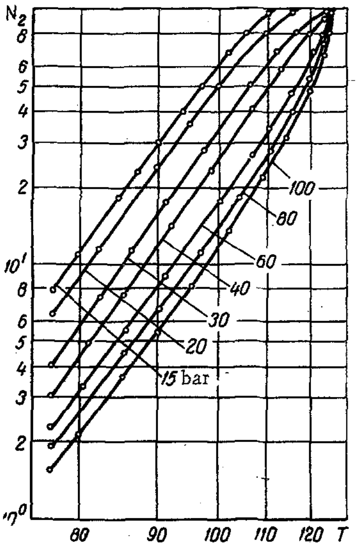

Fig. 3

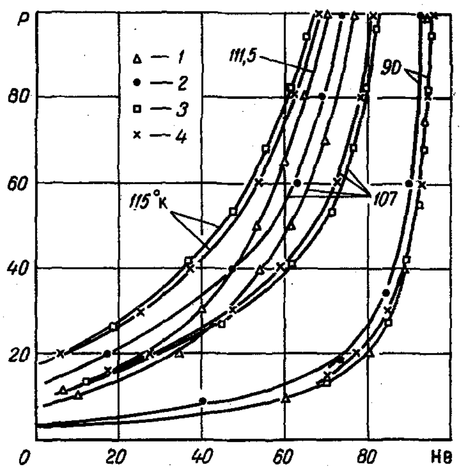

Fig. 4

Fig. 3. Equilibrium composition of N_2–He mixture (%) versus temperature (°K).

Fig. 4. Isotherms of the system vapor–liquid of a N_2–He mixture (P, bar; He, %): 1) data from [3]; 2) from [2]; 3) from [4]; 4) data from present investigation.

obtained for liquid nitrogen are just the same as for the water–air system. These observations later were proposed as the basis for measuring the temperature at the surface of the condensate film.

The change of temperature over the height of the apparatus was monitored and was undertaken in five sections, separated by the same distance from one another.

Observations on the change of composition of the mixture were effected by means of an ITR-1 interferometer and a KhL-6 chromatograph, into which the mixture was fed from the appropriate sections (analysis valves A_1–A_5). In order to ensure a constant composition of the starting mixture in each series of experiments, a closed cycle circuit was used.

When determining the equilibrium compositions of the vapor–gas phase of the nitrogen–helium mixture, static and dynamic methods of investigation were used, for which connecting pieces were mounted on the outer surface of the experimental tube, having an internal slotting. The ends of the couplings were soldered. The volume of the end-space (chamber) between the inner surface of the coupling and the outer surface of the tube was approximately 1 cm^3.

In order that the vapor–gas mixture could reach the annular space, and also in order to return the condensate formed to the cavity of the fractionating tube, openings were drilled in it at the level of the lower end of a coupling. A capillary tube with an external diameter of 0.25 mm was mounted near the upper end of the coupling piece, through a drill hole on the opposite side (for sampling the vapor phase from the chamber).

The outside surface of the coupling piece (from the side of the nitrogen boiling at atmospheric pressure) was insulated by a glass fibre mat and sheathed with a polyethylene film which was impermeable to the liquid.

The small size of the chamber permitted stratification of the mixture to be avoided and by connecting the chamber with the intertube space, a constant pressure was ensured even in those cases when sampling was taking place.

It was established experimentally that the composition of the samples withdrawn remained constant during prolonged opening of the analysis valves. This confirms that the vapor–gas mixture entering the chamber from the main stream and passing through the annular slit with a very low flow rate succeeds in attaining a state which, at a given temperature and pressure, is the equilibrium state. Five chambers were mounted along the height of the experimental tube, separated by the same distance from one another.

TABLE 1. Composition of N_2–He Mixture for Static and Dynamic Conditions

T, °K	Y, % N_2		T, °K	Y, % N_2		T, °K	Y, %N_2	
	static	dynamic		static	dynamic		static	dynamic
	$P = 100$ bar			$P = 40$ bar			$P = 20$ bar	
126,0	100,0	100,0	126,0	100,0	100,0	116,0	100,0	100,0
123,0	63,7	65,5	123,0	88,1	89,2	109,5	80,8	81,9
119,3	44,8	46,5	119,3	76,6	78,1	98,0	43,57	46,67
109,5	22,4	24,2	109,4	47,2	49,0	90,0	24,0	26,7
98,0	9,96	11,2	98,0	21,8	25,0	86,0	17,1	20,0
90,0	5,4	5,70	90,0	11,69	12,89	79,0	7,92	9,32
86,0	3,85	4,2	86,0	8,20	8,7	77,0	6,27	6,60
79,0	1,94	1,98	79,0	3,78	4,0			
77,0	1,57	1,59	77,0	3,0				

Samples were taken from the core of the stream through capillary tubes with a diameter of 0.25 mm from the five corresponding sections.

The equilibrium data, obtained by withdrawing samples from the core of the stream and from the chamber at the appropriate P and T valves, have trivial discrepancies (Table 1) which at elevated and high temperatures become even less. This is explained by the fact that with constant feed the flow rate decreases with increase of pressure of the mixture, i.e. the conditions in the core of the stream become closer to static.

The nature of the change of equilibrium composition of a nitrogen–helium mixture, according to the data from the present investigation, is shown in Fig. 3, as a function of temperature at various pressures, where the equilibrium composition of the vapor (%) is plotted along the vertical axis and the temperature (°K) is plotted along the horizontal axis. Values of the equilibrium concentration obtained for static conditions (in the chambers) are used here.

Comparison of our experimental data with the results of [2-4] is shown in Fig. 4. Here, the pressure of the mixture and the equilibrium composition of the vapor are plotted along the vertical and horizontal axes respectively. It can be seen from the figure that the values of the equilibrium parameters differ considerably between themselves at temperatures above 90°K. Good agreement of the results is observed at all temperatures for the experimental points of this present investigation, with the data from [4].

The maximum error occurring in our experiments, taking into account errors of the measurements of temperatures, concentrations and pressures, does not exceed 1%.

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