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DIFFUSION OF BARIUM VAPOR IN HELIUM

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The diffusion of barium vapor in helium has been studied experimentally by the Stefan method. The pressure values for saturated barium vapor are revised and the diffusion coefficients in a Ba-He mixture are determined.

The present work is devoted to experimental determination of the diffusion coefficient of barium vapor in helium in the temperature range 1180–1420 K at atmospheric pressure (barium vapor is practically monatomic under such conditions) and to revision of the pressure values of saturated barium vapor. In the experiments we used an experimental setup implementing the Stefan method that is close to that described earlier in [1]. For the experiments with barium a 54.5-mm-high diffusion tube (crucible) with an inner diameter of 14 mm was used that was made of tantalum in order to avoid chemical interaction with barium. The tube with barium beneath an inert medium was placed in an evaporator (container) installed in a heated furnace. The furnace was manufactured from ultralight-weight brick and had four Silit electric heaters connected to the heat-regulation circuit. The container was blown with helium, grade A, additionally purified in a special unit filled with activated carbon and placed in a Dewar flask with liquid nitrogen. Helium flowed past the open upper end of the diffusion tube in the container. The partial pressure of barium vapor in helium (at a total pressure close to atmospheric) changed over the tube height from the pressure of saturated vapor near the surface of the melted barium to the low, practically zero, pressure at the upper end. A low vapor pressure in the absence of a marked vortex (a necessary condition in the Stefan method) was ensured by choosing optimal gas flow rates. For helium flow rates from 200 to 300 liter/h the evaporation rate did not change (a weak vortex effect), and at the same time high dilution of the evaporated berium with helium occurred. The container was kept in the furnace for 2–5 h. The temperature was maintained constant within an accuracy of ±1.5% by a BPRT-1 precision temperature regulator.

Experiment duration, temperature, total pressure, and distance from the metal surface to the upper end of the diffusion tube were measured, and mass loss of the metal (tens and hundreds of milligram) was determined by weighing before and after the experiment on an analytical balance with an accuracy of 0.01 mg.

We conducted additional experiments to determine the mass of the metal evaporated upon heating the diffusion tube up to the experimental temperature and in the final stage of cooling. Even in high-temperature experiments the corresponding metal loss did not exceed 10%. In the Stefan method the diffusion coefficient is determined from the experimentally measured evaporation rate (mass loss Δm in the experiment time τ) using the formula

$$PD_{12} = \frac{RTh}{M_1}\frac{\Delta m}{\tau S}\left[-\frac{1}{\ln\left(1-\frac{p_0}{P}\right)}\right] \tag{1}$$

provided $p_0 = p_s$ at the evaporation surface and $p_h = 0$ at the height h at the upper end of the tube.

To process the experimental data on diffusion by this formula it is necessary to have the temperature dependence of the pressure of saturated vapor p_s. However for barium (unlike many other metals) the available temperature dependences of p_s [2–7] show large discrepancies (up to threefold for the vapor pressure). Therefore in processing the experimental results listed in Table 1, we carried out the procedure in reverse order. The values

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TABLE 1. Experimental Results of Diffusion Investigation in a Barium-Helium Mixture

T, K	PD_{12}, N/sec (calculation)	p_s, mm Hg (experiment)	p_s, mm Hg (approximation)	PD_{12}, N/sec (experiment)	d
1177	32.6	0.48	0.51	31.0	0.95
1195	33.5	0.70	0.66	35.7	1.06
1217	34.7	0.89	0.91	33.9	0.98
1240	35.9	1.43	1.24	41.2	1.15
1250	36.5	1.31	1.42	33.7	0.92
1264	37.3	1.47	1.72	31.8	0.85
1290	38.7	2.54	2.40	39.8	1.03
1290	38.7	2.47	2.40	41.0	1.06
1291	38.8	2.29	2.40	36.8	0.95
1318	40.3	3.51	3.37	41.9	1.04
1325	40.7	3.42	3.66	38.0	0.93
1338	41.5	4.45	4.27	43.2	1.04
1355	42.5	5.30	5.16	43.5	1.02
1380	43.9	6.55	6.79	42.4	0.97
1403	45.3	9.76	8,76	50.5	1.11
1422	46.5	9.56	10.61	41.9	0.90

of p_s for barium were reconstructed by processing the experimental data using the temperature dependence of the diffusion coefficient calculated by means of the quantum-mechanical exchange theory of perturbations (ETP) [8].

In the first approximation of the Enskog-Chapman theory, PD_{12} values (in N/sec) are expressed by the formula [9]

$$PD_{12} = 0.0266 \sqrt{T^3 (M_1 + M_2)/(2M_1M_2)} / Q_{12}^{(1.1)}, \tag{2}$$

where $Q_{12}^{(1,1)}$ is the diffusion cross section (Å^2), which depends on the temperature and the potential energy of molecular (in our case barium and helium atoms) interaction. This energy was calculated in [8] using one of the ETP variants [10]. Since the potential well was small (several degrees in temperature units), the results of the quantum-mechanical calculations were approximated by the Born potential:

$$\varphi(r) = B \exp(-r/\rho).$$

The subsequent calculation of $Q_{12}^{(1,1)}$ was performed with the aid of tables [11]. The results of calculations of PD_{11} by formula (2) may be approximated by the relation

$$PD_{12} = PD_{12}^{0} (T/T_{\text{ref}})^n, \tag{3}$$

where $PD_{12}^0 = 38.7$ N/sec at the chosen reference temperature $T_{\text{ref}} = 1290$ K, $n = 1.88$ for the Ba-He mixture investigated. The second column of the table lists PD_{12} values corresponding to relation (3).

The reconstructed p_s values for barium are given in the third column. The corresponding approximate dependence obtained by statistical processing in the temperature range 1180–1420 K with the confidence level 0.95 is as follows

$$\log p_s = (7.37 \pm 0.36) - \frac{9020 \pm 460}{T}. \tag{4}$$

The relative standard deviation for p_s values is approximately 9%. The reconstructed p_s values (the fourth column of the table) lie between the results of [5] and [6]. Finally, the experimental data on diffusion were processed once more by formula (1) using averaged dependence (4) for p_s. The scatter of the PD_{12} values did not exceed ±15% at a relative standard deviation of about 8%.

NOTATION

D_{12}, mutual diffusion coefficient of the binary gaseous mixture components; P, total pressure of the binary gaseous mixture; T, absolute temperature; R, universal gas constant; M_1, atomic weight of the metal; S, cross-sectional area of the diffusion tube; M_2, atomic weight of the gas; $\varphi(r)$, interaction potential of atoms as a function of the internuclear distance r; B, ρ, parameters of the potential; d, ratio of the experimental and calculated PD_{12} values.

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