

KFK-219

**KERNFORSCHUNGSZENTRUM
KARLSRUHE**

März 1964

KFK 219

Institut für Kernverfahrenstechnik

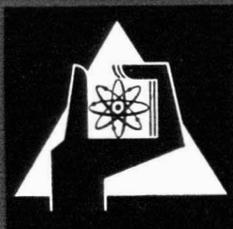
Production of Fast Molecular Beams Using Gaseous Mixtures

R. Klingelhöfer, P. Lohse

Gesellschaft für Kernforschung m. b. H.

Zentrale

7. Okt. 1964



GESELLSCHAFT FÜR KERNFORSCHUNG M. B. H.

KARLSRUHE

Production of Fast Molecular Beams Using Gaseous Mixtures

R. KLINGELHÖFER AND P. LOHSE

Kernforschungszentrum Karlsruhe, Institut für Kernverfahrenstechnik der Technischen Hochschule, Karlsruhe, Germany

(Received 7 May 1963; revised manuscript received 15 October 1963)

According to Becker and Henkes, relatively fast molecular beams of heavy gases can be produced by having a mixture of the heavy gas and a light acceleration gas expand out of a nozzle and transferring the core of the nozzle flow into high vacuum. By mass spectrometric analysis of the collected beam, the degree of enrichment of the heavy gas is investigated for different argon-hydrogen mixtures and for different inlet pressures. The particle flow density of the molecular beam is obtained by measurement of the pressure head. The acceleration of argon is investigated by a time-of-flight method. For an initial mixture of 1% Ar and 99% H₂, the system yields a maximum of 40% for the argon mole fraction in the beam at an inlet pressure of 2 atm. The corresponding flow density is 1.3×10^{17} Ar atoms per cm² per sec. at a distance of 60 mm from the nozzle; the beam velocity is 2100 m/sec corresponding to a kinetic energy of the argon atoms of about 1 eV.

Gesellschaft für Kernforschung m. b. H.
Zentralbücherei 7. Okt. 1964

THE gas kinetic processes on the surface of objects moving at high speed at large altitudes can be simulated in the laboratory by scattering molecular beams of suitable energies from a static test specimen. Thus, for instance, reflection experiments may provide data on the accommodation coefficient which is relevant to aerodynamic drag under the conditions of interest.¹

Becker and Henkes² have shown that high intensity molecular beams in the energy range of interest can be produced by expanding a mixture of a heavy gas and a light acceleration gas out of a nozzle and transferring the core of the nozzle flow into high vacuum. In this way it was possible, for instance, to generate argon molecular beams with a most probable velocity of 2040 m/sec from a mixture of 2.2 mole % Ar and 97.8 mole % H₂ at room temperature. The recorded time-of-flight distributions gave rise² to the conclusion that the light gas used for acceleration strongly separates from the heavy beam gas, thus producing a molecular beam considerably enriched in the heavy component. However, no quantitative data could be given on beam composition and, consequently, on the flow density of the components.

By mass spectrometric analysis of the beam gas, the present work determines the composition of molecular beams produced using Ar-H₂ mixtures under varying conditions. The particle flow density of the beam components is obtained from measurements of the pressure head and the known composition of the beam. The most probable velocity of the beam is determined by the time-of-flight method, as in the earlier work.²

EXPERIMENTAL EQUIPMENT AND EVALUATION OF EXPERIMENTS

The experiments were carried out in a molecular beam apparatus primarily designed for low temperature experiments, this equipment has been described elsewhere.³ In the present work, however, no cooling liquid was used so that the whole beam generating system shown in Fig. 1 was at room temperature. For the maximum inlet pressure used, the pressure in the high vacuum chamber was about 2×10^{-5} mm Hg. Without a beam load, the pressure was about 3×10^{-6} mm Hg.

At a distance of 45 mm from the collimator, the core portion of the beam entered a 33 mm inner diameter tube through a 3.2 mm wide circular opening. The other end of the approximately 1 m long tube was pumped with two cascaded mercury pumps with a capacity of 100 and 12 liter/sec respectively. A trap cooled by liquid nitrogen was located between the tube and the first pump. The gas was collected at the forepressure side of the mercury pumps, and its composition was determined by mass spectrometric means. Calculations showed that under the experimental conditions used there will be no major distortion of the results due to separation effects in the detector. Therefore, with sufficient accuracy, the mole fraction γ_{Ar} as determined in the beam gas can be assumed to be equal to the ratio of the argon particle flow density j_{Ar} to the total particle flow density ($j_{Ar} + j_{H_2}$) of the molecular beam. Thus, the flow density of argon is given by

$$j_{Ar} = \gamma_{Ar}(j_{Ar} + j_{H_2}). \quad (1)$$

¹ See e.g. F. C. Hurlbut, J. Appl. Phys. 28, 844 (1957).

² E. W. Becker and W. Henkes, Z. Physik 146, 320 (1956).

³ E. W. Becker, R. Klingelhöfer, and P. Lohse, Z. Naturforsch. 17a, 432 (1962).

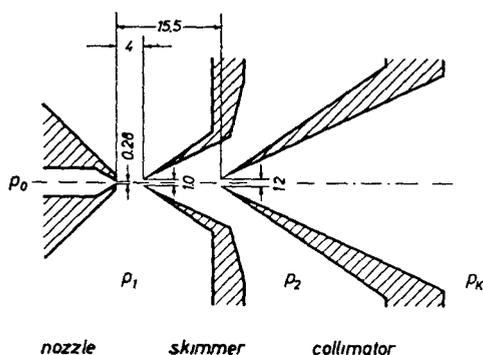


Fig. 1. Beam producing system (dimensions in mm).

The total flow density needed to evaluate Eq. (1) was determined from the pressure head of the beam measured with a collection tube combined with a diaphragm manometer.⁴ For the evaluation of these experiments it could be assumed that the mixture components leave the collection tube under molecular conditions; i.e., with a mean velocity inversely proportional to the square root of their molecular weight. In addition, the molecular beam was investigated with a time-of-flight setup as described in former publications.^{2,5}

DEPENDENCE ON INLET PRESSURE

The upper half of Fig. 2 shows the mole fraction of argon γ_{Ar} of the collected gas as a function of

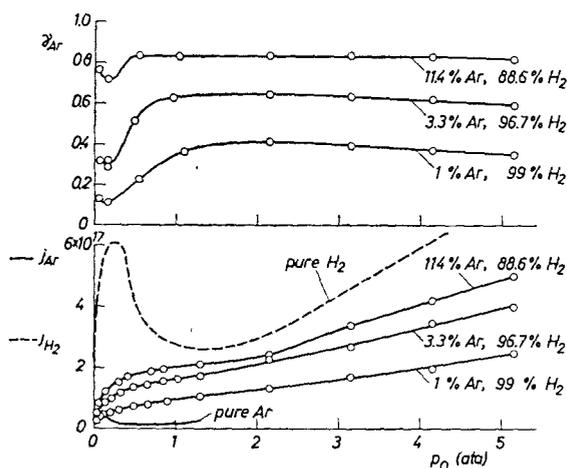


FIG. 2. Mole fraction of argon γ_{Ar} in the collected beam gas and particle flow density j_{Ar} at a distance of 45 mm from the collimator edge as a function of inlet pressure p_0 for three initial mixtures. For comparison, the argon and hydrogen flow densities for pure argon (in units of atoms/cm² sec) and pure hydrogen (in units of molecules/cm² sec) beams are also shown. (1 ata = 735 mm Hg).

⁴ E. W. Becker and K. Bier, *Z. Naturforsch.* 9a, 975 (1954).

⁵ O. Hagen and W. Henkes, *Z. Naturforsch.* 15a, 851 (1960).

inlet pressure p_0 at the nozzle for three different initial mixtures. All curves show a weak minimum at low inlet pressures. γ_{Ar} increases with increasing inlet pressure, in particular for the initial mixtures 1.0 and 3.3 mole % Ar, and reaches a flat maximum at about 2 ata (1 ata = 735 mm Hg). Using the mole fraction of argon in the source gas, $\gamma_{Ar,0}$, one obtains the enrichment factor

$$\beta = \frac{\gamma_{Ar}(1 - \gamma_{Ar,0})}{\gamma_{Ar,0}(1 - \gamma_{Ar})} \quad (2)$$

β reaches maximum values of 69, 54, and 38 for the three mixtures, the initial mixture, having the lowest argon content, providing the largest enrichment factor. The shape of the curves can be explained qualitatively by assuming that the beam producing system acts like a combination of several separation nozzle systems⁶ connected in series, each one reaching the optimum Knudsen number for separation.

The lower half of Fig. 2 shows the dependence of the particle flow density of the argon component on p_0 for the three mixtures investigated. In addition, the argon and hydrogen particle flow densities are given for beams of pure argon and pure hydrogen. The curves for pure argon and pure hydrogen show minimum values at 0.6 and 1.5 ata respectively, which probably are due to the effect of shock waves.⁷ The argon flow density of the seeded beams increases monotonically with inlet pressure for all three mixtures. In the range of mixture ratios investigated, the initial gas with the highest argon content provides the maximum argon flow density in the beam. However, even for the mixture containing only 1% Ar, the argon flow density is still considerably higher than that for the uncondensed beam of pure argon.

The time-of-flight distributions show that for the investigated mixtures at inlet pressures exceeding about 600 mm Hg the velocity⁸ v_m is nearly independent of inlet pressure. The observed values of v_m and the corresponding kinetic energy E_m of the Ar atoms have been compiled in Table I together with the beam velocity w_{exp} , calculated from the measured time-of-flight distributions under the

⁶ E. W. Becker, K. Bier, and H. Burghoff, *Z. Naturforsch.* 10a, 565 (1955); E. W. Becker, K. Bier, and W. Bier, *Z. Naturforsch.* 17a, 778 (1962).

⁷ K. Bier and O. Hagen, *Z. Angew. Physik* 14, 658 (1962). With pure argon above 2 ata there is a partial condensation of the beam gas recognizable by the existence of a second peak in the time-of-flight diagram. Cf also E. W. Becker, K. Bier, and W. Henkes, *Z. Physik* 146, 333 (1956).

⁸ v_m is the velocity corresponding to the maximum of the ion current distribution recorded by the detector of the time-of-flight set up.

TABLE I. Observed values of v_m and the corresponding kinetic energy F_m of the Ar atoms.

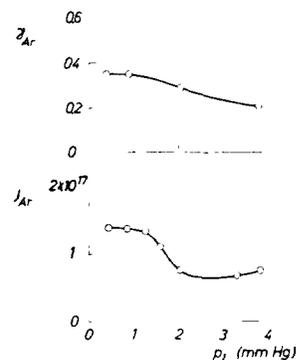
Initial gas mixture	p_0 ata	γ_{Ar}	j_{Ar} atoms/cm ² sec	v_m m/sec	E_m eV	w_{exp} m/sec	w_{theor} m/sec
1 % Ar; 99 % H ₂	3.15	0.40	1.7×10^{17}	2280	1.1	2130	2260
3.3 % Ar; 96.7 % H ₂	3.15	0.64	2.7×10^{17}	1990	0.8	1810	1940
11.4 % Ar; 88.6 % H ₂	3.15	0.83	3.4×10^{17}	1420	0.4	1310	1380
pure Ar	0.16	1.0	0.36×10^{17}	570	0.07	553	550
pure H ₂	0.40	2600	0.07	2460	2460

assumption of a Laval velocity distribution.⁵ For comparison, Table I also includes the velocity w_{theor} of the beam particles which is obtained by assuming a complete transformation of the initial enthalpy of the gas to kinetic energy of directed motion, which of course, in practice is not the case. In the paper quoted above, the results of time-of-flight measurements were already published for a number of argon-hydrogen mixture ratios and inlet pressures up to about 100 mm Hg using a similar beam producing system.

DEPENDENCE ON PRESSURE IN THE SKIMMER AND COLLIMATOR SPACE

In order to investigate the dependence of the mole fraction of the argon in the beam gas and the flow density of argon on the pressures p_1 and p_2 (see Fig. 1), the appropriate pumps were throttled by valves. These experiments were carried out with the mixture consisting of 1% Ar and 99% H₂ at an inlet pressure p_0 of 2.15 ata. Figure 3 shows that γ_{Ar} , as well as j_{Ar} , remained constant within the accuracy of the measurements, when the pressure p_1 between the nozzle and skimmer was increased to about 1.4 mm Hg; i.e., above a nozzle expansion ratio p_0/p_1 of about 10^3 . With further increase of p_1 , i.e., for further reduction of the nozzle expansion ratio, both quantities decrease. This result is in accord with the fact, that, in supersonic beams, the

Fig. 3. Mole fraction of argon γ_{Ar} in the collected beam gas and particle flow density j_{Ar} (in units of atoms/cm² sec) of the argon component in the beam at a distance of 45 mm from the collimator as a function of pressure p_1 between the nozzle and skimmer for the initial mixture of 1% Ar and 99% H₂ at an inlet pressure p_0 of 2.15 ata.



flow of the core portion of the beam between nozzle and skimmer is unaffected by nozzle discharge pressures p_1 below that corresponding to a "critical" expansion ratio.⁷ An increase of pressure p_2 between the skimmer and collimator from 10^{-3} to 10^{-2} mm Hg resulted in essentially no change of γ_{Ar} and a 10% decrease of j_{Ar} .

ACKNOWLEDGMENTS

Thanks are due to Professor E. W. Becker for suggesting this work and for many valuable discussions. The authors wish to thank Miss J. Riemschneider and Mr. K. Maurer for help with the experimental work. Moreover, thanks are due to Deutsche Forschungs-gemeinschaft for financial support.