Production of Fast Molecular Beams Using Gaseous Mixtures

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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 \(1.3 \times 10^{17}\) 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.

**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.¹ 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 \(2 \times 10^{-5}\) mm Hg. Without a beam load, the pressure was about \(3 \times 10^{-6}\) 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 \(\gamma_A\), as determined in the beam gas can be assumed to be equal to the ratio of the argon particle flow density \(j_A\), to the total particle flow density \((j_A + j_B)\) of the molecular beam. Thus, the flow density of argon is given by

\[
j_A = \gamma_A (j_A + j_B).
\]

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

DEPENDENCE ON INLET PRESSURE

The upper half of Fig. 2 shows the mole fraction of argon $\gamma_{Ar}$ of the collected gas as a function of inlet pressure $p_0$ at the nozzle for three different initial mixtures. All curves show a weak minimum at low inlet pressures. $\gamma_{Ar}$ 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

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

$\beta$ 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 systems 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. 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 velocity $v_a$ is nearly independent of inlet pressure. The observed values of $v_a$ and the corresponding kinetic energy $E_a$ 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

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

<table>
<thead>
<tr>
<th>Initial gas mixture</th>
<th>$p_0$</th>
<th>$\gamma_{Ar}$</th>
<th>$j_{Ar}$</th>
<th>$v_m$</th>
<th>$E_m$</th>
<th>$w_{theor}$</th>
<th>$w_{therm}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>ata</td>
<td></td>
<td>atoms/cm² sec</td>
<td>m/sec</td>
<td>eV</td>
<td>m/sec</td>
<td>m/sec</td>
</tr>
<tr>
<td>1 % Ar; 99 % H₂</td>
<td>3.15</td>
<td>0.40</td>
<td>1.7 $\times 10^{14}$</td>
<td>2280</td>
<td>1.1</td>
<td>2130</td>
<td>2260</td>
</tr>
<tr>
<td>3.3 % Ar; 96.7 % H₂</td>
<td>3.15</td>
<td>0.64</td>
<td>2.7 $\times 10^{12}$</td>
<td>1900</td>
<td>0.8</td>
<td>1810</td>
<td>1940</td>
</tr>
<tr>
<td>11.4 % Ar; 88.6 % H₂</td>
<td>3.15</td>
<td>0.83</td>
<td>3.4 $\times 10^{12}$</td>
<td>1420</td>
<td>0.4</td>
<td>1310</td>
<td>1380</td>
</tr>
<tr>
<td>pure Ar</td>
<td>0.16</td>
<td>1.0</td>
<td>0.36 $\times 10^{17}$</td>
<td>570</td>
<td>0.07</td>
<td>553</td>
<td>550</td>
</tr>
<tr>
<td>pure H₂</td>
<td>0.40</td>
<td></td>
<td></td>
<td>2600</td>
<td>0.07</td>
<td>2460</td>
<td>2460</td>
</tr>
</tbody>
</table>

assumption of a Laval velocity distribution. 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 $\gamma_{Ar}$, 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 $10^{4}$. 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 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 $\gamma_{Ar}$ and a 10% decrease of $j_{Ar}$.

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