and reference beams was used. Leith and Upatnieks[4] as well as Lohman[5] have described a technique which makes possible holography on more general and non-microscopic objects. This same technique was used to perform holography without the use of a lens on small objects by Leith and Upatnieks[6]. In experiments which we have carried out on more general objects, the optimum lower level of detectability was about 12μ when holography was performed without a lens. However, the experimental difficulties of obtaining a resolution of about 5μ are very serious. We have used a microscope (objective lens) to record holographically microscopic images of a general character. Although the attractive feature of 'lens-less' imagery was not used, the ability to record a substantial depth, as well as phase information in the object, has been preserved.

Fig. 1 shows a schematic diagram of the experimental arrangement. Fig. 2 shows a reconstruction of a stained specimen of neurones. The difference between Fig. 2a and 2b is that they are derived from the same hologram, but in planes at different depths. In the specimen this difference in depth corresponds to a de-focusing of 40μ. The arrow in Fig. 2b points to two fibres that have a diameter of about 1μ. In both reconstructions a set of concentric ring sections appear which result from multiple reflexions in the optics producing the reference beam.

The hologram was produced on Eastman Kodak '649-F' spectroscopic plate. The exposure time was 20 sec with a 4 mW output from a helium-neon laser.

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Half-lives of Barium-137m, Silver-109m and Rhodium-106 measured with Calculable Overall Accuracy

A FEW years ago, Wasserman, Twardock and Comar[1] investigated the metabolic dissociation of caesium-137–barium-137m in animals. A value for the half-life of barium-137m could be estimated from their data; it appeared to be about 2·52 min, compared with the 2·60 min previously measured by Mitchell and Peacock[2].

By separating daughter from parent by cation-exchange elution and using the ratio-method of measurement[3], an accurate value for the half-life of barium-137m has now been determined with a known degree of contaminational uncertainty. In this respect, any long-lived caesium-137 in the sample introduces virtually no bias into the measured half-life, despite the fact that it increases the background counting rate. Using a crystal of sodium iodide, several mutually consistent series, each comprising fifteen to thirty single measurements, gave the average value $t_{\frac{1}{2}} = 2·557 \pm 0·005$ min, where the stated standard deviation covers the over-all contaminational uncertainty involved. The standard deviation excluding contaminational uncertainty is lower by a factor of three, as shown in Table 1.

The presence of a contaminant with a half-life, $t_{\frac{1}{2}}^*$ (differing from $t_{\frac{1}{2}}$), must induce a bias on the measured value. When the ratio-method is used and the measurements are duplicated with samples 2^n times stronger at the moment

of production, but $n \times t_{\frac{1}{2}}$ min older at the start of the counting period, the bias, b, on the mean, $T_{\frac{1}{2}}$, of fresh and aged samples is given by:

$$b = \frac{2^{n-n*} + 1}{2^{n-n*} - 1} \tfrac{1}{2}\Delta \tag{1}$$

where Δ denotes the apparent increase in half-life due to ageing the sample for $n \times t_{\frac{1}{2}}$ min, and $n* = (n \times t_{\frac{1}{2}})/t_{\frac{1}{2}}^*$.

Denoting the standard deviation by σ, we have $\sigma_{T_{\frac{1}{2}}} = \tfrac{1}{2}\sigma_\Delta$, whence, according to equation (1):

$$\sigma_b = 3\sigma_{T_{\frac{1}{2}}} \text{ for } \left| n - n* \right| = 1 \tag{2}$$

For barium-137m, Δ was found to be nil. I have made the arbitrary assumption that a null value of Δ indicates that $T_{\frac{1}{2}}$ is unbiased by any contaminant(s) for which $\left| n - n* \right| \geqslant 1$. As can be seen from equation (2), this criterion of radio-purity only increases the over-all uncertainty of $t_{\frac{1}{2}}$ (through σ_b) by a limited factor of three. Concerning the possible contaminants for which $\left| n - n* \right| < 1$, a thorough search through all known nuclides[4] disclosed only a few that were of sufficiently long-lived parentage to survive the year-long ageing of the caesium-137 used. Samples of barium-137m were carefully examined for these possible contaminants, using a γ-spectrometer, and in each case the contaminants were found to be absent, that is, so small an upper limit to their amounts could be set that the contribution to the uncertainty of $t_{\frac{1}{2}}$ was negligible.

A chain of genetically related contaminants might, conceivably, make $\Delta = 0$, and yet induce a bias on $T_{\frac{1}{2}}$. However, all significant chains would include α-emitters, and careful scannings of nuclear emulsions, into part of which samples of barium-137m were absorbed, revealed no α-tracks above background. After checking the method with uranium-treated emulsions, an upper limit to chain-contamination was established and was found to be negligibly small.

In the case of silver-109m, which was separated from cadmium-109 by a modification of Sunderman's method of isotopic exchange[5], the possibility of chain-contamination was disregarded since the parent was not a fission-product like caesium-137. Otherwise, the investigation of radio-purity was quite analogous to that already described.

In the case of rhodium-106, ruthenium-106 material purified by Glendenin's distillation procedure[6] was re-distilled, and it was found feasible to trap ruthenium tetroxide in cold 70 per cent perchloric acid, which meant that a sequence of rhodium-106 samples could be prepared by successively re-cycling parental material through the still (taking up the distillational residue in a counting container resembling a slightly V-shaped pipette with capillary stems). Some ruthenium-103 was probably present in the parental material, but rhodium-103m could not be detected by the stilbene crystal used. No trend was seen in thirty non-stop measurements of half-life based on thirty redistillations of a single portion of parental material and, consequently, contaminational uncertainty was assumed to be negligible in comparison with the uncertainty involved in extrapolating the observed values of $t_{\frac{1}{2}}$ to the point of zero dead-time loss of counts.

Table 1

Nuclide	Present value $t_{\frac{1}{2}}$ (sec)	Standard deviation excluding contaminational uncertainty (sec)	Standard deviation including over-all contaminational uncertainty (sec)	Previous value (ref. 7) $t_{\frac{1}{2}}$ (sec)
Barium-137m	153·4	0·1	0·3	156
Silver-109m	39·80	0·06	0·19	40
Rhodium-106	30·36	0·15	0·15	30

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Flux Relationships for Diffusion in Microcapillaries

IT is well known that flux ratios in small capillaries, that is in capillaries where the Knudsen number (N_k, defined as the ratio of capillary diameter to the mean free path of the diffusing molecule) is much less than one, vary as the square root of the inverse molecular weight ratio for binary systems. It has been shown[1] that for counter-diffusion in an 'open' system, corresponding to the physical situation in most transport experiments of the Wicke–Kallenbach type[2], flux ratio may vary from dependence on the square root to the first power of the inverse molecular weight ratio for the bulk diffusion mechanism alone. For capillaries of a fixed length, the progression from square root to first power dependence is associated with increasing Knudsen numbers, the latter mechanism corresponding to $N_k \gg 1$. It is clear, then, that at $N_k \ll 1$ the predominant mechanism of transport is that of Knudsen diffusion, and flux ratios are equal to the square root of inverse molecular weight ratios, and at $N_k \gg 1$ the predominant mechanism of transport is that of bulk diffusion. Flux ratios for sufficiently large N_k will then be equal to the first power of the inverse molecular weight ratios (provided capillary length–diameter ratios do not approach the order of unity).

In the transition region, $0.01 < N_k < 100$, where both types of transport are of importance, it is not known whether a range of capillaries of larger diameter might be included in which under some circumstances the flux ratio for Knudsen transport (always the square root proportionality) would differ from that for the bulk mechanism (tending toward the first power proportionality for sufficiently large diameters). The overall flux ratio would depend on the relative contribution of each mechanism to the overall transport process.

We resolve this problem by determining, under specified conditions, the fraction of total momentum transfer within the capillary due to molecule-wall interactions. This fraction represents the probability, ψ, that a molecule in transit will interact with the wall. In pure Knudsen diffusion all momentum transport is due to molecule wall collisions, and this probability is unity. Conversely, for the purely bulk transport mechanism this probability is essentially zero, and in the transition region $0 < \psi < 1$. A simple momentum balance for binary diffusion under isothermal, isobaric conditions in the capillary yields:

$$N_A = \left[\frac{1}{D_A} + \frac{1 - y\alpha}{\mathcal{D}_{AB}} \right]^{-1} \nabla C_A \qquad (1)$$

in which N_A is the flux of component A in moles/time-area, D_A the Knudsen diffusion coefficient for A within the capillary, $\mathcal{D}_{AB}$ the bulk diffusivity of the A–B pair, y the mole fraction of A, C_A the concentration of A in moles/volume and α defined as $(1 + N_B/N_A)$. The bracketed term in this equation is proportional to total momentum

transfer. On integration over total capillary length for unity Δy, it may be shown that ψ as obtained from the ratio of the Knudsen contribution to the total is:

$$\psi = \left(\frac{1}{N_k \alpha} \right) \ln \left\{ \frac{N_k + 1}{N_k (1 - \alpha) + 1} \right\} \qquad (2)$$

If it is desired to define the transition region in terms of set limits on ψ, as above, then equation (2) provides the means for computing corresponding capillary dimensions under specified conditions of temperature and pressure. For a given value of ψ, the capillary diameter corresponding is directly proportional to temperature and inversely proportional to pressure. Capillary dimensions corresponding to the upper bound of transition zone diffusion (at $\psi = 0.1$, for example) are of the same order of magnitude as those reported as characteristic of many porous structures, particularly those used in catalysis (with the exception of very high pressure conditions). These diameters range generally from 1000 Å to 10,000 Å, and transition region diffusion is thus probably the predominating transport mechanism in such structures.

Flux ratios for counter-diffusion in the transition region can be determined as a linear combination of flux ratio for pure Knudsen diffusion, R_k, and that for the bulk process, R_B, under the conditions of operation. The probability determined from equation (2) is utilized in this computation:

$$R_T = \psi R_k + (1 - \psi) R_B = 1/(\alpha - 1) \qquad (3)$$

in which R_T is the observed flux relationship for the combined process. R_k is, of course, always equal to the square root of inverse molecular weight ratio, and the proportionality of R_B to the inverse molecular weight ratio is determined by methods outlined previously[1]. This is a trial-and-error solution, since ψ depends on the flux ratio, as specified by equation (2).

Figs. 1 and 2 show the results of such computations for the helium–argon (I) and hydrogen–argon (II) systems, which are typical of those used in the study of diffusion through porous structures. The computations are based on an argon mean free path of 1100 Å at 300° K and one atmosphere, and, in Fig. 2, a capillary diameter of 2000 Å, and $L/D = 50$, which is an appropriate average value for catalyst support structures as discussed below. Fig. 1 demonstrates the form of ψ in terms of the Knudsen number as a function of the overall flux ratio, R_T (equation (2)). In Fig. 2 it is seen that as diameter is varied the deviation of α (or R_T) from the square root of inverse molecular weight ratio becomes apparent only for low values of (L/D). Our investigations of porous structures[3,4], however, indicate that characteristic values of (L/D) are normally of the order of 50–100, and one would expect to observe, in general, no deviations. For (L/D) of these magnitudes, indeed, further computation indicates that flux ratio deviations would never become apparent, other
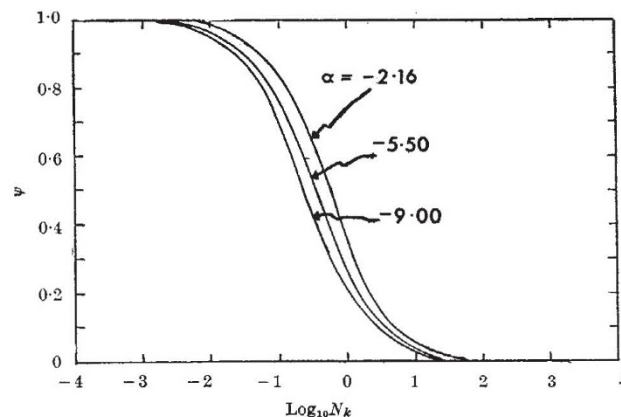

Fig. 1. Fraction of momentum transport by molecule-wall interactions: helium–argon system (equation 2)