Open-ended Plastic Capillary Method for Diffusion Experiments with β-active Tracers

Self-diffusion of Cl^- in Aqueous NaCl

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An open-ended capillary method, where the capillary is drilled directly into a plastic scintillator, is described. The method is tested with ^{36}Cl in 0.1 M and 0.5 M NaCl solutions.

The open-ended capillary method first applied by Anderson and Saddington[1] has frequently been used with γ-active tracers, e. g. ^{22}Na[2], but also with β-active tracers, e. g. ^{36}Cl[3–7], where the monitoring is more difficult. Gosman[4] applied a continuous monitoring method for ^{36}Cl, but the glass capillaries, he used absorbed much of the β-activity, making the measurements unsatisfactory.

So we decided to try a new method. As a basis we used a capillary which was drilled directly into the plastic scintillator, as has already been proposed by Mills and Godbole[8]. The drilled plastic capillary is, of course, not as uniform as a fabricated glass capillary, but – as the authors have shown[9] – irregularities in the capillary can be taken account of in the calibration procedure.

We tested the method by measuring the self-diffusion coefficient of Cl^--ion in two NaCl solutions. The results agree with earlier data measured with the diaphragm method.

Experimental

Figure 1 is a schematic diagram of the measuring apparatus. Figure 2 shows the experimental counting efficiency function, which was measured using a $Ag^{36}Cl^-$ point source while the capillary was filled with water.

The scintillator (diam. 25.4 mm, length ~50 mm) was made of two pieces of plastic scintillator (NE 102A, Nuclear Enterprise). The capillary (diam. 1 mm) was drilled into one of them which was then joined to the other with optical cement (Ne 580). The effective length of the capillary was (25.38 ± 0.02) mm. The outer surfaces of the scintillator, except that against the photomultiplier and a small area around the mouth of the capillary, were painted with reflector paint (NE 560). The scintillator was connected to the photomultiplier (EMI 9524 B) with a high grade silicon oil.

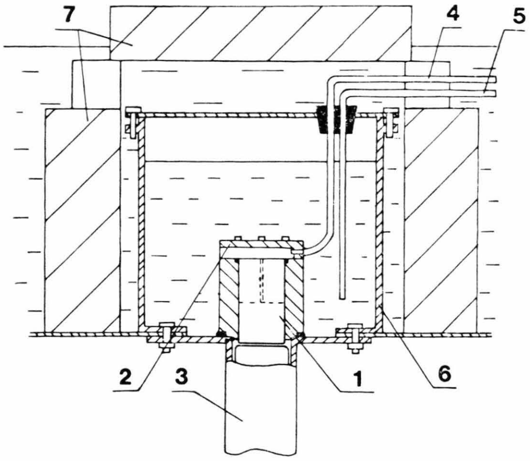

Fig. 1. Schematic diagram of the apparatus. 1 Scintillator with teflon case; 2 teflon cover with a 5×5 mm^2 flow channel; 3 photomultiplier and steel case; 4 inlet tube; 5 outlet tube; 6 vessel of the outer solution; 7 lead bricks.

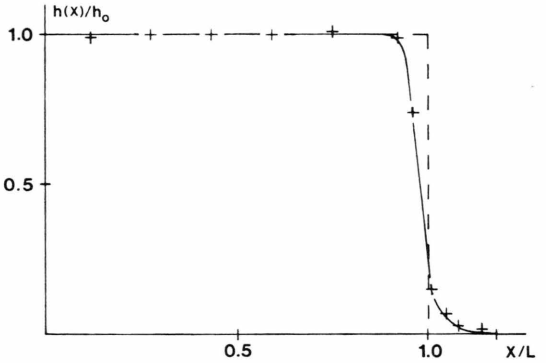

Fig. 2. The relative counting efficiency function. h_0 is the pulse count with the source at the bottom of the capillary of length L.

In the experiments Merck's PA grade NaCl and a $Na^{36}Cl$ solution with a specific activity greater than 3 mCi/gCl supplied by the Radiochemical Centre were used. The water was triple distilled, and all the solutions were made by weighing, the accuracy being better than 1%. The activity of the capillary solution (volume about 20 μl) was usually between 500 – 700 counts/s at the beginning of the experiments.

The experimental procedure was as follows: The capillary was filled with the active solution so that there remained a small drop at the top of the capillary, while the outer vessel was partly filled with

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the inactive solution. The teflon cover was then fastened with screws and the pumping of the inactive solution was started. Altogether about 1000 ml of the inactive solution, were used, part of which being in a bottle serving as a stabilizing vessel of the flow. The temperature was (298.15 ± 0.02) K.

The flow rate of the outer solution was (2.0 ± 0.1) mms^{-1}, which with the self-diffusion coefficient $1.952 \times 10^{-9}\ \text{m}^2\,\text{s}^{-1}$ of Cl^- in 0.1 M NaCl[2] resulted in an effective capillary length of 25.38 mm. Instead, with the rate (1.0 ± 0.1) mms^{-1}, e. g., the effective length of the capillary was 25.83 mm (Δl-effect!).

Results and Discussion

The self-diffusion coefficient of Cl^--ion was calculated by the method of least-squares using the equation

$$\ln I(t) = \ln I_0 - \pi^2 D\, t/4\, L^2\,,$$

where $I(t)$ is the activity of the capillary solution at time t, I_0 a constant, L the length of the capillary, and D the diffusion coefficient. The activity $I(t)$, corrected against the background, was actually the pulse count in the time interval Δt divided by Δt[10]. In the experiments Δt was usually 2700 s and the time t used for the evaluation of D was between 8.6×10^4 s ... 2.6×10^5 s.

We performed experiments with two NaCl solutions. With a 0.1 M solution we made a calibration, the mean value of D in three experiments having been $(1.953 \pm 0.003) \times 10^{-9}\ \text{m}^2\,\text{s}^{-1}$. We then tested the apparatus with a 0.5 M solution with the result $D = (1.848 \pm 0.003) \times 10^{-9}\ \text{m}^2\,\text{s}^{-1}$. Mills[2] has given the value $(1.854 \pm 0.009 \times 10^{-9}\ \text{m}^2\,\text{s}^{-1}$, which was measured with the diaphragm method. In previous experiments with the openended capillary the results have not been as accurate[2, 3].

Although the results agree very well with the generally accepted data one must, nevertheless, pay attention to a possible source of systematic error, i. e. to the Δl-effect.

One way to avoid the Δl-effect is to use a closed capillary. This type of method is under development in our laboratory.

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