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Automatic counting of fission fragments tracks using the gas permeation technique

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Abstract

An automatic counting system for fission tracks induced in a polycarbonate plastic Makrofol KG (10 μm thickness) is described. The method is based on the gas transport mechanism proposed by Knudsen, where the gas permeability for a porous membrane is expected to be directly related to its track density. In this work, nitrogen permeabilities for several Makrofol films, with different fission track densities, have been measured using an adequate gas permeation system. The fission tracks were produced by irradiating Makrofol foils with a 252Cf calibrated source in a 2π geometry. A calibration curve fission track number versus nitrogen permeability has been obtained, for track densities higher than 1000/cm^2, where the spark gap technique and the visual methods employing a microscope, are not appropriate for track counting.

Keywords: Fission tracks; Gas flow; Measuring instruments

1. Introduction

The presence of natural and artificial radioactive substances in the environment has, over the past decades, been of great concern to the general public and is the subject of many scientific researches. Among the studied radioactive pollutants some attention has been given to the actinide nuclei due to their long physical half-lives and high biological toxicity [1–3].

Fission track registration techniques have been widely used to determine the concentration of these fissile nuclides in environmental as well as in biological samples [2–5]. The densities of fission fragment tracks recorded in an etched polycarbonate foil have been determined by visual methods employing optical microscopes, by processing the track images with an image analyser as well as by automatic spark gap chambers. However, visual methods are in general tedious and time consuming and the image analyzers coupled to PC computers have the inherent limitation of the instrument image definition and thus, they are recommended only for counting of low track densities. In addition, spark gap chambers are able to count track densities up to approximately 1000 tracks/cm^2. Above this limit there is a counting saturation due to the spark superposition on the chamber electrodes. Consequently, there is a need for the

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development of new counting techniques to cover the region of high track densities.

The main objective of the present work was to apply the gas permeation technique for determining fission tracks registered in polycarbonate plastics, as an effort to offer a new track counting tool mainly to reach the region of high track densities.

Various transport mechanisms of gases through porous membranes have been presented in the literature depending on the structure of the membranes. The main mechanisms reported are the Surface, Knudsen and Poseuille diffusion [6–8]. For pore sizes smaller than 100 nm and at low pressures, as is the case of the present experiment, Knudsen diffusion is the predominant transport mechanism.

The gas flux density through a membrane per unit area and per unit time is defined by [6,8]

$$J = P\Delta p \tag{1}$$

where J is the gas flux density (mol s^{-1} m^{-2}), P (mol s^{-1} m^{-2} Pa^{-1}) the permeability and Δp is the pressure difference (Pa) across the membrane.

According to Knudsen theory, the permeability is given by [9]

$$P = P_k = \frac{2\varepsilon\mu_k\, vr}{3RT\delta} \tag{2}$$

where ε is the porosity and equal to $n\pi r^2$, n is the number of pores per m^2, $r(m)$ the mean pore radius of the membrane, μ_k the shape factor which is equal to unity for uniform parallel straight pores normal to the surface of the membrane, R the gas constant (8.314 J mol^{-1} K^{-1}), T the absolute temperature (K), δ the thickness of the membrane and v the average velocity (m s^{-1}) of the gas which is equal to $(8RT/\pi M)^{1/2}$, where M being the molecular mass of the gas (kg mol^{-1}).

The permeability is obtained by the angular coefficient of the straight line fitted to the experimental data of the curve flux density versus transmembrane pressure. According to Eq. (2) one can observe that P is directly related to the membrane track density n.

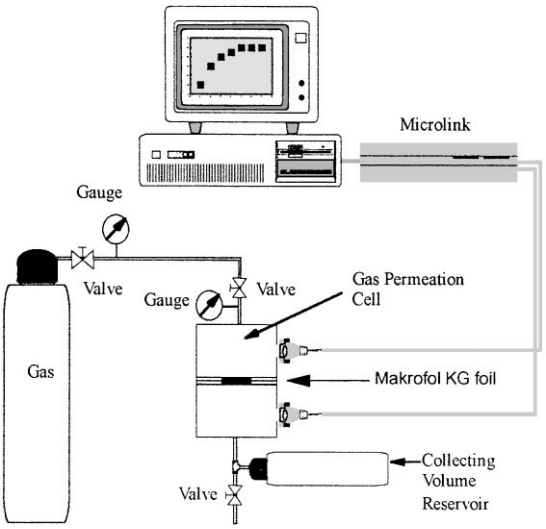

Fig. 1. Schematic diagram of the gas permeation system.

2. Experimental

The gas permeation system [10] presented in Fig. 1 was used for determination of gas permeabilities through the plastic membranes. The polycarbonate plastic Makrofol KG, thickness 10 μm, manufactured by Bayer Chemicals, Germany and the nitrogen gas have been used in the present work. The equipment consisted of a pressurised nitrogen gas cylinder, a pressure regulator, a gas permeation cell, a pressure transducer and a computer link for data logging.

Samples with different track densities were produced by irradiating Makrofol KG foils in a calibrated fission source of 252Cf for different exposition times. The 252Cf source activity was (477.9 ± 1.8%) Bq on April 27, 1993. All Makrofol foils were simultaneously etched in a 35% KOH solution at $T = 60°C$, for 325 s in order to obtain tracks with diameters around 100 nm. After the etching, Makrofol KG samples with 10 mm diameter were fixed between the input and output compartments of the permeation cell for the determination of the respective nitrogen permeabilities. The interface of the cell compartments was sealed using Nescofilm tape. This cell allows to apply a gas pressure on one side of the film (high-pressure

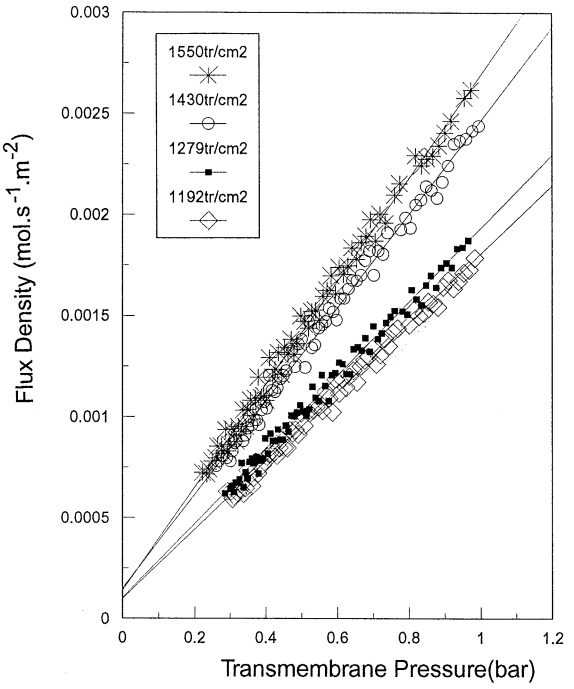

Fig. 2. Linear dependence of the flux density with the transmembrane, pressure.

compartment) and to collect the gas passing through it into a well-known storage volume (low pressure compartment) on the other side. By monitoring the gas pressure in both compartments of the permeation cell as a function of the time, the nitrogen permeabilities could be determined from the slope of the curves of gas flow J in the steady state, versus transmembrane pressure differences Δp.

3. Results and Discussions

Fig. 2 shows as an example, the linear dependence of the nitrogen flux density with the transmembrane pressure obtained for Makrofol detectors with track densities of 1550, 1430, 1279 and 1192 tracks/cm^2.

The reproducibility of this method was not so good (around 10%) and it was the most important source of experimental error considered in this experiment. Additional experimental studies are being carried out in order to reduce this error source.

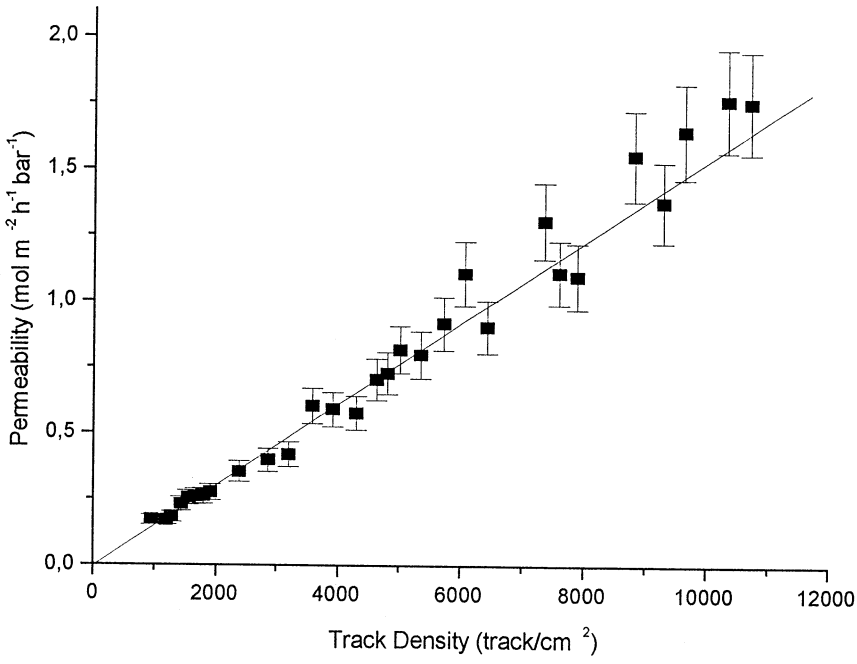

Fig. 3. Nitrogen permeability versus track density for the Makrofol KG fission track detector.

The nitrogen permeabilities obtained, plotted against the fission track densities in the interval from 954 to 10 726 tracks/cm^2, are presented in Fig. 3. The best fitting to the calibration data points obtained by using the least-squares method [11] was a straight line $y = a + bx$ where $a = -(3 +/ -12) \times 10^{-3}$, $b = (1.536 +/ - 0.053) \times 10^{-4}$.

The normalized χ^2 was obtained as 0.6 showing the excellent quality of the fit. Using this calibration curve, Makrofol foils can have the track densities determined, with an overall error ranging from 3% to 6%, within the interval considered. The higher the track density, the lower the error.

4. Conclusion

A new technique for fission track counting has been developed in this work using the methodology of gas permeation through porous membranes. It is relatively rapid (some minutes) if an appropriated collecting storage volume is used, inexpensive and can in principle be applied for counting of any track density. In the present experiment it was tested employing the polycarbonate plastic Makrofol KG, of 10 μm thickness, with track densities ranging from around 1000 to 10 000 tracks/cm^2.

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