

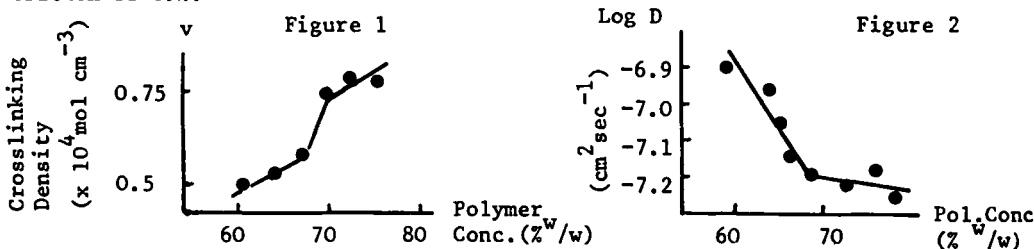
## INFLUENCE OF GEL STRUCTURE ON DIFFUSION THROUGH POLY(2-HYDROXYETHYL METHACRYLATE (polyHEMA) GELS

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Two important factors which affect diffusion characteristics of solutes through hydrogels are the crosslinking density of the polymer and the organisation of water within the gel. The crosslinking density,  $v$ , can be calculated from the results of stress-strain tests on the polymer, while the crosslinking density of an ideal network can be written as  $v_c = 2c$  where  $c$  is the concentration of a difunctional crosslinking agent, such as ethylene glycol dimethacrylate (EGDMA). Discrepancies between  $v$  and  $v_c$  arise from incomplete reaction of the crosslinking agent, chain entanglements and non-covalent molecular interactions between the polymer chains. In this work,  $v$  and  $v_c$  have been measured for polyHEMA gels and the results correlated with values of the diffusion coefficient of a solute through the gels. Diffusion coefficients were measured using the double-disc method (Park and Van Hoang 1979).

The tensile tests were carried out using a J.J. Lloyd tensile tester, type 22K. Gels in the shape of dumbbells were prepared by  $\gamma$  irradiation of the monomer solution (HEMA and EGDMA in water) in a suitable mould, under nitrogen. The samples were elongated at a fixed rate of strain on the instrument under conditions of 100% relative humidity.

The tensile properties of gels of different crosslinker and polymer concentration were measured, and values of  $v$  and  $v_c$  calculated. A plot of  $v$  against  $v_c$  was linear and gave a value of  $0.5 \text{ mol cm}^{-3}$  for the physical crosslinking of the network, where  $v_c = 0$ . The slope was 0.82, indicating the crosslinking reaction is approximately 80% efficient. A plot of  $v$  against polymer concentration (Fig. 1) was also linear but showed a sharp displacement at a polymer concentration of 69%.



A plot of log diffusion coefficient of salicylic acid vs polymer concentration in the same gels (Fig.2) also showed a change of slope at a polymer concentration of 69%. This concentration corresponds approximately with the limit of freely diffusing or bulk water in the gel. In gels of higher polymer concentration, only interfacial and bound water exist (Andrade et al 1975). Since it is thought that in gels of lower polymer concentration, transport is by pore flow through the bulk water, it appears that at a hydration of 31%, the small pore size and absence of bulk water preclude pore flow and transport is thereafter by a dissolution and diffusion mechanism through the polymer chains. The sharp increase in crosslinking density at this point supports this view and indicates that the intermolecular bonding in the network is increased by the absence of freely diffusing water in the network. Therefore, in formulating a controlled release implant, it appears desirable to formulate the gel to have a high value for  $v$  since small changes in hydration of such gels (e.g. from imbibing body fluid) will not change the diffusion coefficient significantly.

Andrade, J.D., Jhon, M.S. and Lee, H.B. *J. Coll. Interface Sci.* 51, 225 (1975)  
Park, G.S. and Hoang, Tran Van, *Eur. Polym. J.* 15, 817 (1979)