

COMMUNICATIONS

Casting solvent effects on the permeability of polymer films of differing quaternary ammonium (cation) content

R. S. OKOR*, W. ANDERSON, *Department of Pharmaceutics, University of Strathclyde, Glasgow G1 1XW, UK*

Films of certain acrylate-methacrylate copolymers of varying cation (quaternary ammonium) content were cast from three different solvents: acetone, chloroform, and ethanol; their permeability to urea was determined at 30 °C. In films containing 66 mole cation mol⁻¹ polymer chain, and cast from acetone, chloroform or ethanol, the permeation rates were 480, 900 and 2807 mg h⁻¹, respectively. Differences in permeation rates were however less significant when polymers of cation content lower than 46 mole cation mol⁻¹ polymer chain were used to form films. This observation may relate to differences in the degree of dissociation of the polymer in the different solvents.

Various studies (Lehmann 1968; Okor 1982a, b) have indicated the potential of films of certain acrylate-methacrylate copolymers in controlled release and dialysis applications. Mass transport through these films occurs through water-filled pores in the film structure (Abdel-Aziz et al 1975). Variation in the quaternary ammonium (cation) content in the film-forming polymer (Okor 1982a) and composition of the casting solvent (Abdel-Aziz et al 1974) affect the porous structure and hence the permeability of the resulting films. In that study, mixtures of acetone and ethanol in varying proportions were used as casting solvents. Since the solvent composition at the gelation stage will not necessarily be the same as the initial composition (partial pressures of the component solvents being different), it would be more appropriate to carry out the study using pure solvents if film property is to be related to the solvent property. Therefore, in the present study pure solvents acetone, chloroform, and ethanol have been used for casting films of polymers which differ only in the cation content, with a view to establish any possible effect of the solvent type on film permeability.

Method

Two acrylate-methacrylate copolymers, designated A and B were received under the trade names Eudragit RL100 and RS100, respectively, from Rhom Pharma Darmstadt; the polymers differ only in their content of quaternary ammonium (cation) groups in a ratio 2:1. A had 66 moles cation per mole of polymer chain whilst B had 33 moles per mole of polymer chain. The cation content was determined by measuring the total nitrogen in a given weight of polymer (Okor 1982a).

* Correspondence and present address: Department of Pharmaceutics, University of Benin, Benin City, Nigeria.

Polymers of intermediate cation content and hydrophilic (polar) character were produced by mixing A and B in varying proportions in the casting solvent. Glycerol triacetate (reagent grade, BDH) was used as plasticizer in film casting. Chloroform, acetone (both Analar), and ethanol (absolute alcohol) were obtained from BDH and used as casting solvents. Urea (Analar grade, 10% w/v solution in water) was used as solute permeant.

Single films, 19.8 ± 0.3 μm thick were cast from an organic solution (solvent: acetone, chloroform, or ethanol) containing 2.5% w/w of the polymer, and 0.5% w/w of glycerol triacetate as plasticizer using the casting technique of Abdel-Aziz et al (1975). Resulting films were placed in a desiccator (charged with dried silica gel) 24 h before their evaluation.

Urea permeation rate through the films (exposed film area 23 cm²) was measured using the method of Abdel-Aziz et al (1975). Films of varying polymer cation content were used. With 135 mL of 10% urea solution in the donor compartment and 135 mL water in the acceptor compartment, the permeation experiment was conducted at 30 ± 0.05 °C with stirring of compartment fluid at 300 rev min⁻¹ with magnetic stirrers. The film lower surface (in contact with substrate during film formation) was upstream in all permeation experiments. The amount of urea appearing in the acceptor fluid was determined spectrophotometrically (Watt & Chrisp 1954) at 1 hourly intervals for 5 h. Each experiment was carried out in 4 replicates and the results were used to calculate the mean permeation rate; results were reproducible to ±6.5% of the mean.

Results

Amounts of urea appearing in the acceptor compartment as a function of time are shown in Fig. 1; films of polymer A were used in this experiment. Permeation rates (obtained from the slopes of the linear portions of these plots) were 480, 900 and 2807 mg h⁻¹ for acetone-, chloroform-, and ethanol-cast films, respectively. As can be seen in Table 1 these differences are only marked in films with high polymer cation content >46 mole cation mol⁻¹ polymer chain, below this cation level differences in the permeability of the three types of films are generally less significant.

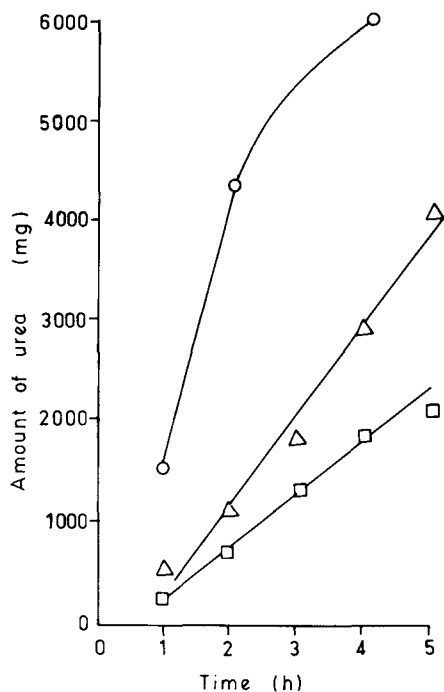


FIG. 1. Amount of urea permeating the film of polymer A as a function of time. The films were cast from different solvents: acetone \square , chloroform \triangle , and ethanol \circ .

Discussion

Solvents may influence the porous structure of cast films by their high volatility and hence rapid desolvation of the film to a highly porous structure or by the solvent interaction with polymer chains to influence polymer-polymer cohesion. In the present study the first factor may be considered unimportant since the more volatile solvents, acetone and chloroform, produced less permeable films compared with the ethanol-cast films. In an earlier study (Okor 1982a) it was shown that film permeability was strongly dependent on polymer cation content with permeability increasing about 500-fold when cation content was increased 2-fold from 33 to 66

Table 1. Permeability of films cast from different solvents: Acetone (F-ac), chloroform (F-ch), and ethanol (F-et).

Film type of copolymer	Cation content, mol mol ⁻¹ polymer chain	Urea permeation rates, mg h ⁻¹ in films:		
		F-ac	F-ch	F-et
A	66	480	900	2807
A0.8*	59	157	240	1094
A0.6	53	53	28	962
A0.4	46	35	21	34
A0.2	40	30	8	13
B	33	1	8	9

* Fraction of polymer A in mixture of polymers A and B.

moles cation mol⁻¹ polymer chain, an effect which was associated with strong polymer cohesion at lower cation levels. In the present study such strong cohesion is considered dominant over the effects of casting solvent which is expected to be minimal because of the lower cation groups available for interaction with the solvents, hence at this cation level there were lower differences in permeability of films cast from the different solvents. The exact nature of the cation-solvent interaction is not immediately obvious, but it may involve changes in the degree of dissociation in the different solvent systems. For instance, a higher degree of dissociation in ethanol to furnish more charged groups (cation) in the polymer structure is expected to lead to greater mutual repulsiveness of polymer chains (Okor 1982a), resulting in a more porous and permeable film; these aspects however require further studies.

REFERENCES

- Abdel-Aziz, S. A. M., Armstrong, P. A. M., Anderson, W. (1974) *J. Pharm. Pharmacol.* 26: Suppl. 131P-132P
 Abdel-Aziz, S. A. M., Anderson, W., Armstrong, P. (1975) *J. Appl. Polym. Sci.* 19: 1181-1192
 Lehmann, K. (1968) *Drugs made in Germany* 11: 34-36
 Okor, R. S. (1982a) *J. Pharm. Pharmacol.* 34: 83-86
 Okor, R. S. (1982b) *Int. J. Pharmaceutics* 11: 1-9
 Watt, G. W., Chrisp, J. D. (1954) *Anal. Chem.* 26: 452-454