

IJP 03231

Water vapour permeation of aqueous based ethylacrylate methylmethacrylate copolymer films

L. Baert and J.P. Remon

Laboratory of Pharmaceutical Technology, University of Gent, Harelbekestraat 72, 9000 Gent (Belgium)

(Received 14 February 1992)

(Accepted 1 May 1992)

Key words: Water vapor permeation; Isolated film; Eudragit NE 30 D; Fick's law; Polyvinylpyrrolidone; Permeability

Summary

The water vapour permeation rate of isolated films consisting of Eudragit NE 30 D was found to be dependent on the film composition, film thickness, isolation technique and thermal treatment. The temperature during the experiments appeared to influence dramatically the water vapour permeation rate. With increasing temperature, the water permeation rate of the film increased. It was shown that Fick's law was not obeyed for water vapour permeation of aqueous based Eudragit NE 30 D films, indicating that investigating different film compositions at one thickness leads to erroneous conclusions.

Introduction

This study reports on the water vapour permeation of pure Eudragit NE 30 D films, Eudragit NE 30 D-polyvinylpyrrolidone and Eudragit NE 30 D- α -lactose monohydrate films. Eudragit NE 30 D is a film forming material that is frequently used in controlled release formulations. The films were prepared by casting or by roll casting. The influence of thermal treatment was investigated. Water vapour permeation rates were calculated and related to film thickness. It was shown that Fick's law was not applicable for water vapour

permeation in the case of films containing Eudragit NE 30 D.

Materials and Methods

Composition of the different films

Three film compositions were used: (Formula 1) Eudragit NE 30 D; (formula 2) Eudragit NE 30 D, polyvinylpyrrolidone, water (100:4:72, by wt); (formula 3) Eudragit NE 30 D, α -lactose monohydrate 200 M, water (100:6:72, by wt). Eudragit NE 30 D (Röhm Pharma GmbH, Weiterstadt, Germany) is an aqueous polymeric dispersion of a neutral copolymer of ethylacrylate and methylacrylate and has a dry weight of 28.5–31.5%. α -Lactose monohydrate 200 M (Pharmatose 200) and polyvinylpyrrolidone (Kollidon 30) were purchased from De Melkindustrie Veghel, Veghel,

Correspondence to: J.P. Remon, Laboratory of Pharmaceutical Technology, University of Gent, Harelbekestraat 72, B-9000 Gent, Belgium.

The Netherlands and BASF, Ludwigshafen, Germany, respectively.

Isolation and treatment of the films

Casting The dispersions were cast on a glass plate using the method described by Banker et al. (1966). After drying in a room at 25% R.H. and 22°C for 8 h the glass plates were kept at 4°C for 1 h. Next the films were isolated and fixed on a paper sheet at room temperature. Half of the films were thermally treated at 40°C for 6 h. Water vapour permeation experiments were performed 1 day after isolation.

Roll casting The aqueous polymeric dispersions were poured in a recipient and a glass cylinder rotated in the dispersion as depicted in Fig. 1. The rotational speed was set at 1 rotation

per 3 min. The diameter of the glass roll was 15 cm and the length 40 cm. Roll casting was performed at 25% R.H. and 22°C. After drying, the film was isolated from the glass cylinder and half of the film surface was thermally treated as described above.

Determination of film thickness

The thickness of the films was measured using an Elcometer 256 F (Elcometer Instruments Ltd, Manchester, U.K.). The film was placed on an iron surface. A hard polymeric film (Folex[®]) of about 100 μm was placed on top of the film. This hard polymeric film prevented the deformation of the film during thickness determination. The apparatus was calibrated with calibration films (Elcometer Instruments Ltd, Manchester, U.K.)

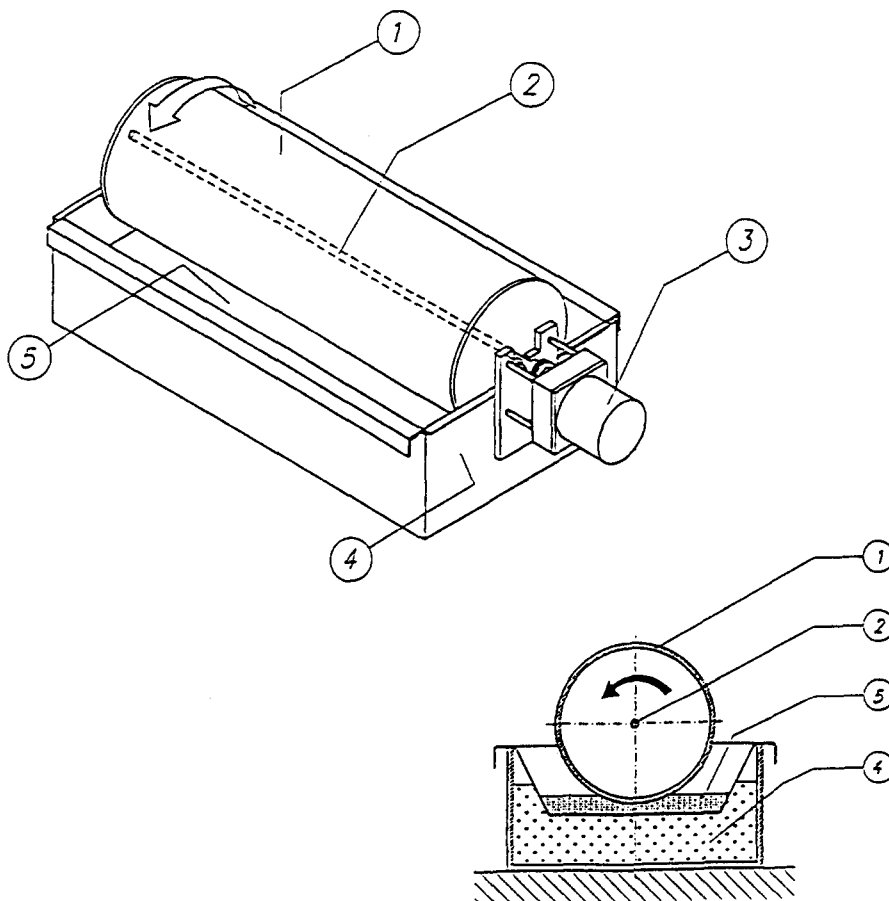


Fig. 1. Schematic view of the roll caster. 1, roller; 2, axis; 3, motor; 4, fluid container kept at a certain temperature; 5, dispersion.

of 50.7 and 123 μm , respectively. The thickness was measured at 10 different points randomly chosen on a surface of 4.91 cm^2 .

Water vapour permeation

The permeation cell used was described by Patel et al. (1964). The volume of the cell was adapted to 100 ml and the inner diameter on the top was 25 mm. The polymeric film was placed so that the film side sticking to the glass plate during the isolation procedure was in contact with the inner side of the permeation cell. The permeation cell was filled with a saturated potassium chloride solution creating an atmosphere of 85% R.H. within the cell. The cell was placed in a controlled room at 25% R.H. and a temperature of 22°C (for one composition tests were also performed at 27°C). The permeation cell was weighed at the beginning of the experiment, after 8 h and after 24 h. A linear relationship was observed between time and the amount of water permeated.

The water vapour permeation rate was calculated using the formula $R_{\text{wvp}} = W/A \cdot \Delta p$ where

R_{wvp} is the water vapour permeation rate ($\mu\text{g h}^{-1} \text{cm}^{-2} \text{mmHg}^{-1}$), W the amount of water permeated through the film in $\mu\text{g/h}$, A the area of the exposed film (cm^2) and Δp the vapour pressure difference (mmHg). The permeability was calculated using the formula $P = R_{\text{wvp}} \cdot t$ where t is the thickness of the film.

Results and Discussion

On the basis of Fick's law, the water vapour permeability constant P is defined as $P = W \cdot t/A \cdot \Delta p$ where W is the amount of water diffusing per h ($\mu\text{g/h}$) through a film of thickness t (cm) and area A (cm^2). The vapour pressure difference Δp across the film is expressed as mmHg. The application of Fick's law for water vapour permeation of films has been a matter of controversy in the literature. Kanig et al. (1962) mentioned that Fick's law was not applicable for water vapour permeation of ethylcellulose, methylcellulose, zein and ethylcellulose-polyvinylpyrrolidone films and observed a linear relationship

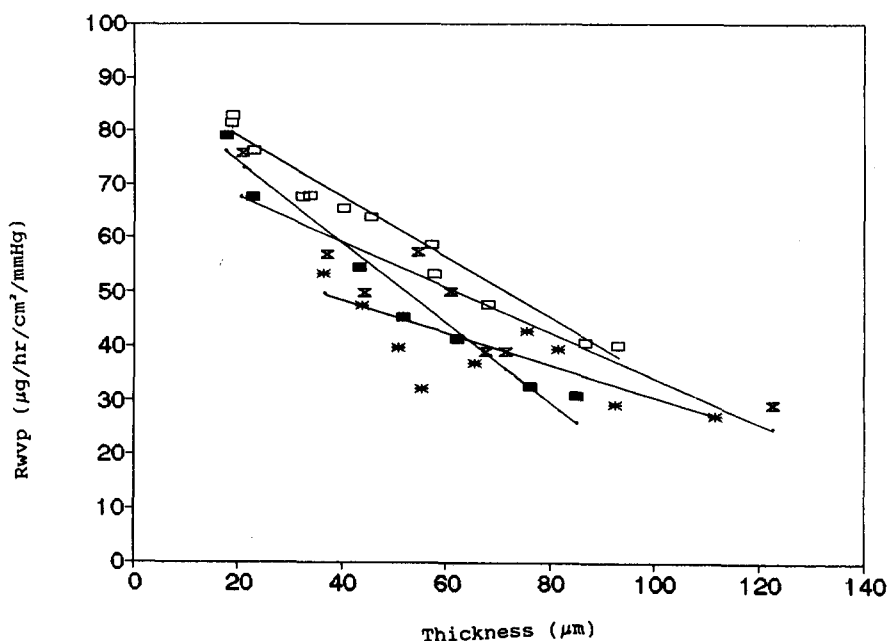


Fig. 2. Water vapour permeation rate ($\mu\text{g h}^{-1} \text{cm}^{-2} \text{mmHg}^{-1}$) as a function of film thickness for films made of pure Eudragit NE 30 D. Cast, non-thermally treated film (\square); cast, thermally treated film ($*$); roll-cast, non-thermally treated film (\blacksquare); roll-cast, thermally treated film (\otimes).

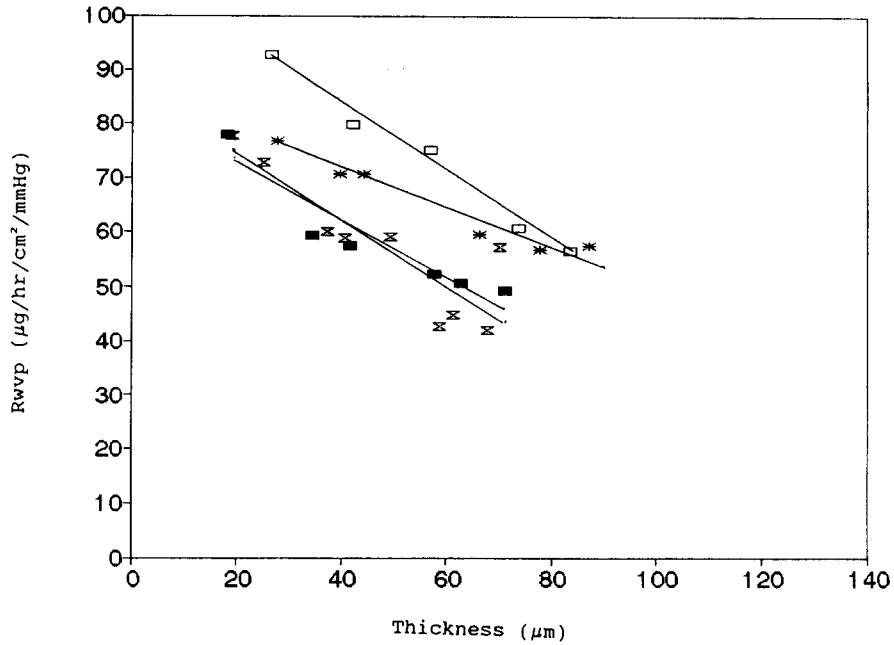


Fig. 3. Water vapour permeation rate ($\mu\text{g h}^{-1} \text{cm}^{-2} \text{mmHg}^{-1}$) as a function of film thickness for films made of Eudragit NE 30 D and polyvinylpyrrolidone (Kollidon K 30). Cast, non-thermally treated film (\square); cast, thermally treated film (*); roll-cast, non-thermally treated film (\blacksquare); roll-cast, thermally treated film (\otimes).

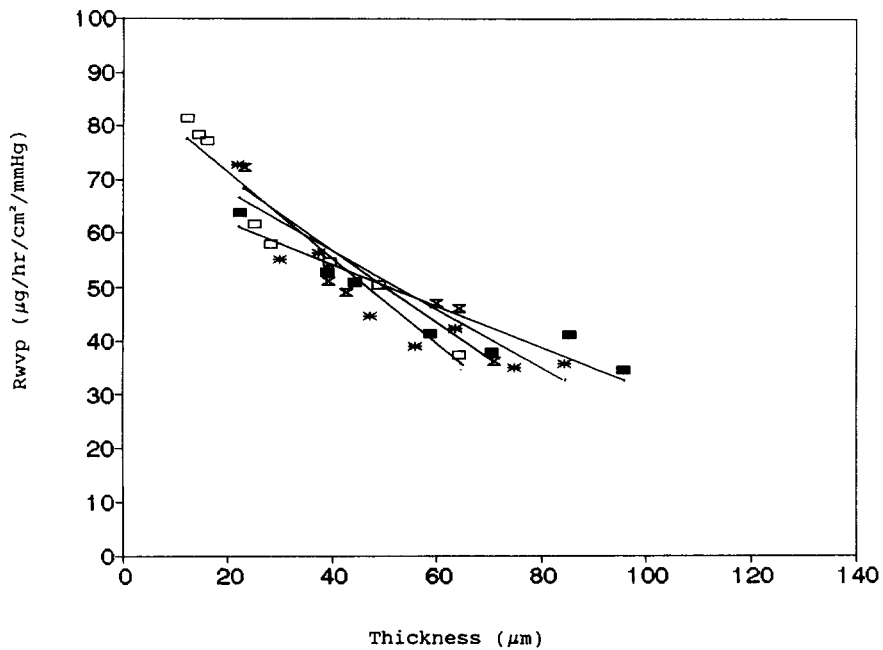


Fig. 4. Water vapour permeation rate ($\mu\text{g h}^{-1} \text{cm}^{-2} \text{mmHg}^{-1}$) as a function of film thickness for films made of Eudragit NE 30 D and lactose. Cast, non-thermally treated film (\square); cast, thermally treated film (*); roll-cast, non-thermally treated film (\blacksquare); roll-cast, thermally treated film (\otimes).

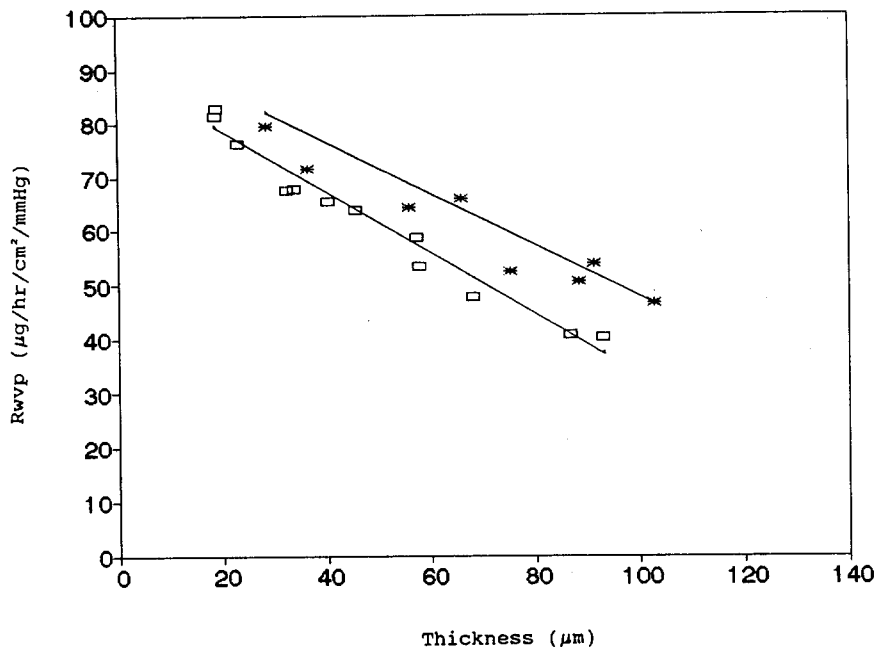


Fig. 5. Water vapour permeation rate ($\mu\text{g h}^{-1} \text{cm}^{-2} \text{mmHg}^{-1}$) as a function of film thickness for films made of pure Eudragit NE 30 D tested at 22°C (\square) and 27°C (*). The films were cast and non-thermally treated.

between water vapour permeation rate and film thickness. Parker et al. (1964) observed the same relationship for hydroxypropylmethylcellulose-low molecular weight ethylcellulose films loaded with talc or titanium dioxide. Patel et al. (1964) proved that the permeability of cellulose acetate propionate, cellulose acetate butyrate and cellulose acetate hydrogen phthalate films was not a constant value as expected from Fick's law while cellulose acetate stearate films approached Fickian behaviour in their permeability to water vapour. An explanation for this phenomenon was that when water absorption was considerable, the gradient across the film was no longer uniform and deviation from Fick's law occurred. Banker et al. (1966) showed that the application of Fick's law to water vapour permeation data was dependent on the type of films and proved that only for lipophilic films (e.g., butylmethacrylate films) was the law applicable.

The results reported by Joshi et al. (1989) might be questionable as the authors used the ASTM designation no. E 96-80 in order to calculate the permeability of hydroxypropylmethyl cellulose films thinner than 1 mm while the ASTM

designation specified a film thickness between 12.5 and 32 mm. Munden et al. (1964) Who investigated more than 30 different film formula-

TABLE 1

Coefficient of the linear equation for the relationship between film thickness and permeability

Formula	Slope	Calculated y-intercept r	
Films tested at 22°C			
1 C NT	-0.5684	89.38	0.9823
1 C T	-0.2694	57.07	0.7798
1 R NT	-0.6914	85.76	0.9795
1 R T	-0.4210	74.87	0.8891
2 C NT	-0.6337	108.91	0.9917
2 C T	-0.3496	85.38	0.9751
2 R NT	-0.4982	81.67	0.9312
2 R T	-0.5945	85.69	0.8656
3 C NT	-0.8104	87.67	0.9653
3 C T	-0.5367	75.43	0.9091
3 R NT	-0.3654	67.71	0.9318
3 R T	-0.6018	80.40	0.9081
Films tested at 27°C			
1 C NT	-0.04203	89.21	0.9605

1-3 correspond to formulae 1-3; c, cast; R, roll-cast; T, thermally treated; NT, non-thermally treated.

tions, Allen et al. (1972) studying cellulose acetate films and Porter et al. (1982) who investigated cellulose acetate phthalate and polyvinyl acetate phthalate films used Fick's law for interpretation of their data without examining the influence of film thickness on permeability.

We investigated whether Fick's law was applicable to Eudragit NE 30 D films by making films of different thickness. Table 1 and Figs 2–4 show a nearly linear relation between the water vapour permeation rate and the thickness of the film. As expected, films were less permeable with increasing thickness. The calculation of the permeability values ($P = W \cdot t / A \cdot \Delta p$) revealed that permeability (P) was not a constant value as would be expected if Fick's law was applicable. Fig. 6 shows an example of the evolution of the permeability (P) of a pure Eudragit NE 30 D film as a function of increasing film thickness.

The method of preparing films had an influence on the permeation rate. In all cases, roll-cast films were more permeable than cast films except for a non-thermally treated Eudragit NE 30 D- α -

lactose monohydrate film. Thermal treatment had an unpredictable influence on the permeation rate. The slope of the permeation rate-film thickness plots for thermally and non-thermally treated films was similar or changed abruptly. In those cases where slope alteration was observed, non-thermally treated films were more permeable at lower film thickness while at higher film thicknesses, thermally treated films showed greater permeability. This indicates that it is irrelevant to study film permeability characteristics based on experiments at one film thickness. The composition of the Eudragit NE 30 D film influenced the permeation rate but no general conclusions could be drawn. Fig. 5 shows the influence of variation in ambient (22 and 27°C) temperature on the permeation rate. Increasing the experimental temperature increased the permeation rate.

In conclusion, it can be said that the water vapour permeation rate of Eudragit NE 30 D films is dependent on the composition, thickness, isolation technique and thermal treatment of the films.

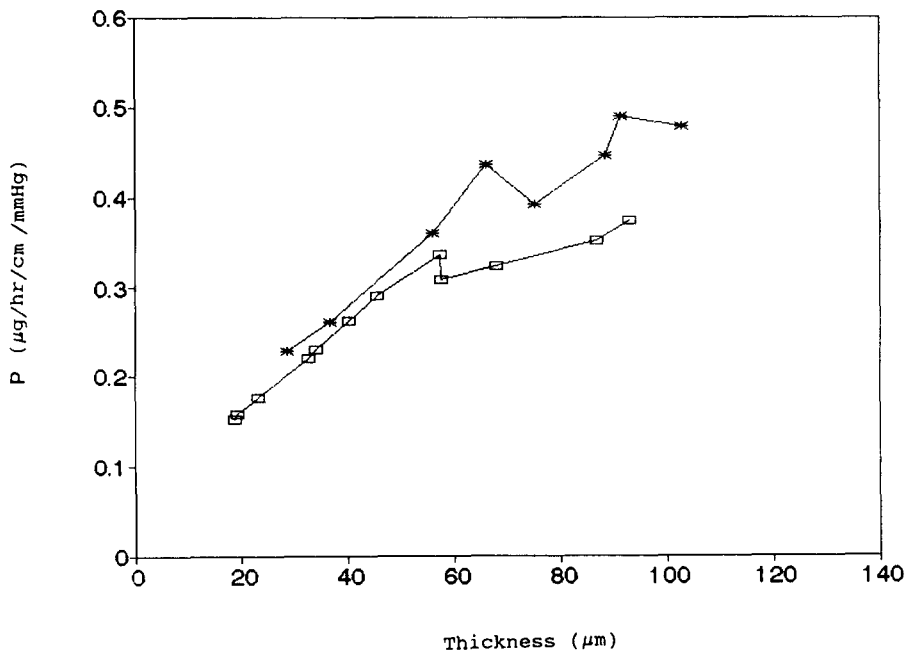


Fig. 6. Permeability ($\mu\text{g h}^{-1} \text{cm}^{-2} \text{mmHg}^{-1}$) as a function of film thickness for films made of Eudragit NE 30 D tested at 22°C (□) and at 27°C (*). The films were cast and non-thermally treated.

Acknowledgements

The authors wish to thank P. De Baets, A. De Latthauwer and R. De Smet for technical assistance and SMB-Technology-Galephar and the IWONL for financial support.

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