# Determination of the Oxygen Transmissibility and Permeability of Hydrogel Contact Lenses

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**ABSTRACT:** To test the validity of the method of stacked hydrogel contact lenses to obtain the oxygen permeability and transmissibility coefficients of the lenses, the coefficients of one low hydration (38% water) and two high hydration (55 and 58% water) hydrogel contact lenses stacked one to five on an oxygen electrode were determined. From the oxygen diffusion through the lenses, the current intensity in the stationary state was determined, and from this the "instrument" the oxygen transmissibility was obtained. The permeability coefficients of the lenses, corrected for edge effects, were obtained from the slope of the plot of the reciprocal of the transmissibility coefficients versus the lens thicknesses. The oxygen permeability and transmissibility coefficients of the lenses obtained neglected the boundary layers resistance between the stacked lenses and, therefore, these are not the "true" coefficients. This article compares the "apparent" oxygen permeability coefficients of the hydrogel contact lenses, obtained by others, with the "true" oxygen permeability coefficients obtained with a corrected equation that takes into account the boundary layers between the stacked lenses. © 1999 John Wiley & Sons, Inc. J Appl Polym Sci 72: 321–327, 1999

**Key words:** oxygen transmissibility; oxygen permeability; oxygen diffusion; hydrogel contact lenses

## INTRODUCTION

The oxygen transmissibility ($Dk/L_{av}$) of contact lenses of average thickness $L_{av}$ and the permeability coefficients ($Dk$) of the material of the construction of the lenses are two of the most important physical parameters of these lenses.[1–5] The thickness of commercial contact lenses, because of their specific optical power, is not uniform from the center to the edge of the lenses. Therefore, for calculation of the oxygen transmissibility of specific lenses, the harmonic average lens thickness ($L_{av}$) of these lenses is determined by the integration of the inverse of the thickness ($1/L$) of $n$ concentric rings of thickness $L_1 \ldots L_n$.[6,7] The harmonic average oxygen transmissibility governs the actual flux of oxygen to the anterior cornea and determines the amount of the corneal swelling under a given lens. The permeability coefficient of a material is the product of its oxygen diffusion coefficient ($D$) and the oxygen solubility coefficient in the material ($k$). The $Dk$ units are usually given in $cm^3$ $O_2$ (STP) $cm/cm^2$ s mmHg.

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But the practical unit for the $Dk$ is the barrer [1 barrer = $10^{-11}$ cm$^3$ $O_2$ (STP) cm/cm$^2$ s mmHg].

Four different procedures have been used to determine the oxygen transmissibility and permeability coefficients of contact lenses. Three of these procedures use a Clark oxygen electrode covered by the lens, directly[8] or separated by a thin Teflon membrane of known oxygen transmissibility,[9,10] to measure the oxygen flux through the lenses. The first method was developed for use with hydrogel lenses that in the hydrate state are swollen in the electrolyte that is used in the electrochemical reaction that takes place on the electrode. The second method is similar to the first, but is adapted for use with rigid hydrophobic contact lenses. In this case, a thin piece of cigarette paper soaked in an electrolyte solution is sandwiched between the hydrophobic lens and the electrode to establish the electrolytic contact between the lens and the electrode.[11–14] The third method, which can be used for hydrogels as well as for rigid lenses, contains the electrolyte solution between the Teflon membrane and the electrode.[9,15] The fourth method uses dual chambers separated by a lens. The oxygen is introduced into one of the chambers and diffuses through the lens from the chamber with the higher partial pressure of oxygen to the second chamber fitted with an oxygen consuming electrode.[16–19]

The electrochemical technique, described by Aiba et al. for polymeric membranes,[20] has been used often with hydrogel contact lenses placed directly on the electrode for the determination of the oxygen permeability coefficient of the lenses.[11–15,21–28] The oxygen flux through the lenses is determined from the measurement of the electric current in a potentiometer. When the gold cathode is maintained at 0.75 V with respect to the silver anode, all the oxygen passing through the sample is reduced at the cathode. For small electric current densities, the nature of the reduction process in the cell varies with the pH of the solution.[28] However, at pH between 5 and 12 (borax buffer), used in most experiments, the following electrochemical reaction takes place at the cathode surface:

$$O_2 \text{ (dissolved)} + 2H_2O + 4e^- \rightarrow 4OH^-$$

At the steady state, the apparent oxygen transmissibility (AOT) is related to the total current diffusion (I, in amperes), by eq. (1)[29]:

$$\frac{Dk}{L_{av}} = \frac{I}{nFA\Delta p} = BI \tag{1}$$

where $n$ = 4 (number of electrons generated at the electrode), $F$ (Faraday constant) = 96,487 Coulomb/mol vol $O_2$ (STP) = 96,487 amperes s/22.4 $\times$ $10^3$ cm$^3$ of $O_2$ (STP), $A$ (area of lens exposed to the gold cathode) = $14.24 \times 10^{-2}$ cm$^2$, and $\Delta p$ ($O_2$ partial pressure difference across the lens at sea level) = 155 mmHg. Therefore, $B = (nFA\Delta p)^{-1}$ = 0.02629 cm$^3$ $O_2$ (STP)/cm$^2$ amperes s cmHg is a constant for the permeability cell in the given conditions.

Knowing the average thickness of the lens ($L_{av}$ in cm), one can obtain the oxygen permeability ($Dk$) of the lens material from eq. (2):

$$Dk = BIL_{av} \tag{2}$$

Equation (3) was developed because the oxygen transmissibility measured according to eq. (1) does not take into account the resistance to the transmission of oxygen by the liquid boundary layers between the lens and the cathode and over the lens:

$$\left(\frac{L_{av}}{Dk}\right)_{app} = \left(\frac{L_{av}}{Dk}\right)_{lens} + \left(\frac{L_{bl}}{D_{bl}k_{bl}}\right) \tag{3}$$

where ($L_{bl}/D_{bl}k_{bl}$) is the boundary layers resistance. Equation (3) has been used by several investigators to determine the oxygen permeability coefficient of hydrogel contact lenses using the so-called stack procedure.[11–13,21,22,25] The stack procedure is an alternative to the measurement of the oxygen flux through a series of lenses of progressive thicknesses to obtain the oxygen permeability coefficient of the contact lenses and the boundary-layer resistance.[10] In the stack procedure, stacked hydrogel contact lenses of the same kind, with equal thickness, optical power, and hydration, are used to obtain the oxygen permeability as a modification of the Aiba et al. procedure described above. Using this technique successively to determine the transmissibility of one lens, followed by the transmissibility of stacks of two to five lenses, the oxygen transmissibility and permeability coefficients of the lenses corrected for edge effects could be obtained from the slope of the plot of the reciprocal of the transmissibility coefficients versus the total thickness of the lenses in each stack. Nevertheless, the permeability obtained with eq. (3) is also an “apparent” permeability, because this equation does not take into account the liquid layers sandwiched between the lenses in the stacks.[21,22] Similar criticism applies to the determination of the oxygen

**Table I Characteristics of Lenses Used in This Study**

| Lens | Material | Manufacturer | Hydration (% $H_2O$) | $L_{av}$[a] ($\mu$m) |
|---|---|---|---|---|
| Seequence | Polymacon[b] | Bausch & Lomb | 38.6 | 116 ± 2 |
| Newvue | Vifilcon A[c] | Ciba Vision | 55.0 | 113 ± 2 |
| Acuvue | Etafilcon A[d] | Johnson & Johnson | 58.0 | 102 ± 2 |

[a] Average harmonic thickness.[32]
[b] Crosslinked poly(2-hydroxyethyl methacrylate).
[c] Crosslinked copolymer of 2-hydroxyethyl methacrylate with methacrylic acid and vinyl pyrrolidone.
[d] Crosslinked copolymer of 2-hydroxyethyl methacrylate with methacrylic acid.

permeability of hydrophobic rigid contact lenses when thin cigarette paper soaked with the electrolyte solution is sandwiched between the lenses and the oxygen electrode.

This article introduces a new equation [see below, eq. (4)], which takes into account all the fluid-boundary-layers' resistance, to obtain the true oxygen transmissibility and permeability coefficients of one low hydration (38% water) and two high hydration (55 and 58% water) hydrogel contact lenses, using the stacked lenses procedure, and compared the results to those obtained by other investigators with eq. (3)[21,22] and with the coefficients reported by the manufacturers of the lenses.

## EXPERIMENTAL

The characteristics of the lenses used in this study are given in Table I. All the lenses had the same optical power (−3.00 D), radius of curvature (8.80 mm), and diameter (14.00 mm). The flux of oxygen through the lenses as a function of time was determined using the electrochemical technique described by Aiba et al.,[20] modified for the determination of the oxygen transmissibility and permeability coefficients of contact lenses.[12–14,21–28]

The experimental procedure used the Clark oxygen electrode to determine electrochemically the oxygen transmissibility of hydrogel lenses according to the ISO/DIS 9913 and 9913-1 standards.[30] The current density at the cathode was measured using a cell that consists of a potentiometer (Schema Versetae, Albany, CA) coupled to a permeometer (Model 201T, Rheder Development Co., Albany, CA). The gold cathode had a surface area $A = (14.24 \pm 0.13)10^{-2}$ cm$^2$. The silver anode was positioned concentrically to the cathode, and these were isolated by an epoxy resin, altogether forming a spherical cap.

All measurements of the current diffusion were performed at 35 ± 1°C, following two procedures. In procedure A, the electrode was wetted with a drop of 0.9% NaCl before the hydrated lens was taken from the supplier bottle and was placed on the surface of the electrode. The lens was gently pressed onto the electrode with a hollow plastic cylinder after placing an O-ring between the lens and the cylinder, taking care not to rupture the lens. Immediately thereafter, about 0.5 cm$^3$ of the salt solution was applied through the cylinder on top of the lens. Because the salt solution and the lens were saturated with atmospheric oxygen, before each experiment and with the current turned on, nitrogen saturated with water vapor was bubbled through the salt solution in the cylinder until the current read on the electrode decreased to near zero. Then, moist air, at 1 atmosphere, was bubbled through the saline solution on top of the lens until the electric current reached the stationary state.

Procedure B was similar to procedure A, except that the initial removal of the dissolved oxygen from the lens and solution was not performed. Thus, the initial partial pressure of oxygen was the partial pressure of oxygen in the atmosphere (155 mmHg). Because, initially, the salt solution on the cathode is in equilibrium with the atmospheric oxygen, the rate of oxygen reduction starts at a higher level and decreases to the stationary state. Procedure A was used only to obtain the apparent diffusion coefficients. Procedure B was used in the rest of the experiments. At the completion of all the measurements, the current versus time records were analyzed, and from the steady-state current intensity values, we calculated the mean values of the AOT and standard deviation, corrected for edge effects, of the lenses.

The harmonic average thickness ($L_{av}$) was calculated from five measurements using an electronic lens thickness gauge,[6a,b] one in each of the five concentric zones of the central area of the lens with radius 2.5 mm, which is equal to the area of a lens in contact with the cathode. The lenses were measured in random order until five readings of each lens had been obtained. Reproducibility of the results to ±2.5% was found in all cases.

Equation (4) incorporates, in addition to the resistance (inverse of the transmissibility) of the boundary layers ($L_{bl}/D_0k_0$), the resistance of the fluid layers sandwiched between the stacked lenses ($L_{sl}/D_0k_0$):

$$\left(\frac{L}{Dk}\right)_{app} = \left(\left(\frac{L_{bl}}{D_0k_0}\right) - \left(\frac{L_{sl}}{D_0k_0}\right)\right) + \mathbf{n}\left(\left(\frac{L_{av}}{Dk}\right) + \left(\frac{L_{sl}}{D_0k_0}\right)\right) \quad (4)$$

where $\mathbf{n}$ is the number of lenses in the stack. $L_{sl}$ and $L_{bl}$ are the thicknesses of the layers sandwiched between the stacked lenses and the boundary layers between the lens and the electrode and on top of the upper most lens, respectively. $D_0k_0$ is the permeability of the fluid layers between the lenses (which is approximately the oxygen permeability of water). The slope of eq. (4) includes the total resistance (inverse of the true transmissibility coefficients) of the lenses and the resistance of the sandwiched fluid layers between the lenses. If we neglect the resistance of the sandwiched liquid layers between the lenses, eq. (4) will be identical to eq. (3). In such a case, from the slope of eq. (3), we can obtain the apparent transmissibility of the lenses, and from the intercept, we can obtain the diffusion resistance of the boundary layers. From the permeability coefficient of the fluid layers (essentially water), we can obtain their overall thickness.[31] Then, from the slope of the plot of $(L_{av}/Dk)_{app}$ versus $\mathbf{n}L_{av}$, we can calculate the oxygen resistance of the lenses, and from the inverse of the resistance, we can obtain the true permeability coefficient of the lenses $(Dk)_{true}$.

Because the transmissibility of the lenses is much higher than that of the very thin aqueous boundary layers, we used eq. (5) from the time-lag method described by Aiba et al.[20] to make an approximate estimation of the apparent diffusion coefficient for the lens– fluid layer system:

$$D = \frac{L^2 I_{st}}{6(I_{st}t_{st} - q_{st})} \quad (5)$$

where $L$ is the total thickness of the lens and the boundary liquid layers; $\mathbf{I}_{st}$, the steady-state current in the stationary state; $t_{st}$, a time large enough for the current to be practically indistinguishable from the stationary current; and $q_{st}$, the total charge which passes through the cathode in the time $t_{st}$. Because the very thin aqueous boundary layers have much higher oxygen transmissibility than that of the lenses, eq. (5) can be used to calculate approximate "apparent" diffusion coefficient values.

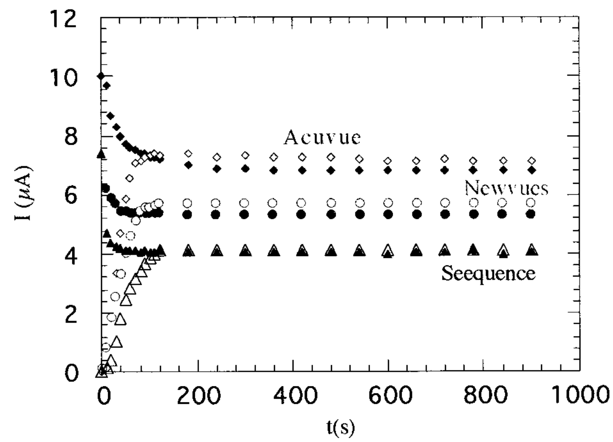


**Figure 1** Time course of the electric current readings with the three kinds of hydrogel contact lenses on the oxygen electrode, obtained using experimental procedure (white) A and (black) B.

## RESULTS AND DISCUSSION

The typical time evolution of the current reduction obtained for Acuvue, Seequence, and Newvue hydrogel lenses are given in Figure 1. A close inspection of this figure permits us say that the steady state and the current reduction were the same with either procedure A or B. The current intensity in the steady state was obtained when the intensity reached a constant value for at least 2 min.

Figures 2–4 show the time evolution of the current reduction for the Acuvue, Seequence, and Newvue lenses (stacking one to five lenses), respectively . The current starts at a high level in all cases, due to the atmospheric oxygen in the system, but decreases rapidly and at 5–6 min reaches the steady state, which is used for the calculation. From eq. (3), we obtained the apparent resistance of the lenses (reciprocal of the apparent transmissibility) $(L_{av}/Dk)_{app}$. Then, the $(L_{av}/Dk)_{app}$ for each stack of one to five lenses is plotted versus the harmonic mean thickness of the lenses as shown in Figure 5 for Acuvue,

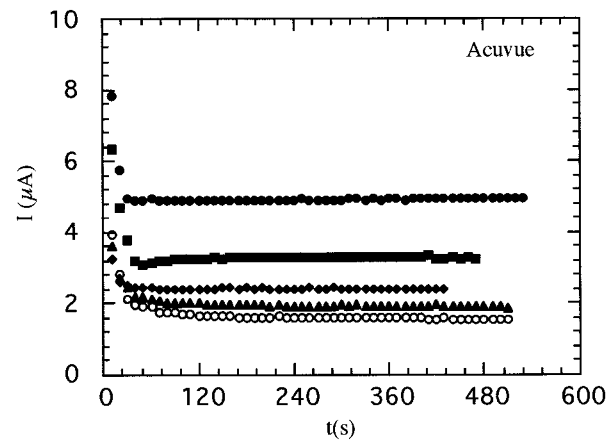


**Figure 2** Time course of the electric current obtained from (●) one, (■) two, (♦) three, (▲) four, and (○) five stacks for the Acuvue lenses on the oxygen electrode.

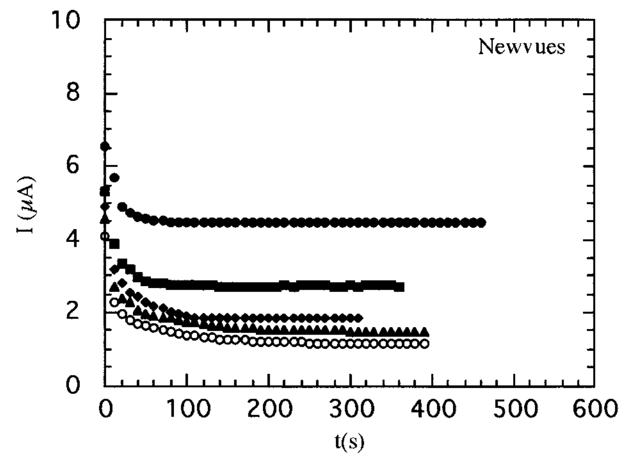


**Figure 4** Time course of the electric current obtained from (●) one, (■) two, (♦) three, (▲) four, and (○) five stacks for the Newvue lenses on the oxygen electrode.

Seequence, and Newvue, respectively. In all cases, the plotted straight lines have correlation coefficients of about 0.99.

The plot of $(L_{av}/Dk)_{app}$ versus the number of lenses per stacks [**n,** eq. (4)] for the three contact lenses is shown in Figure 6, which is used to calculate the AOT and permeability coefficients given in Table II. This table also gives the apparent diffusion coefficient of the lenses derived from eq. (5). If there were no sandwich liquid layers between the stacked lenses, the apparent coefficients (Table II) would be the "true" coefficients for the lenses. However, the "true" permeability coefficient of the contact lenses (Table III) were obtained from the plots [eq. (4)] of the apparent oxygen resistance $[(L_{av}/Dk)_{app}]$ versus the number of lenses in the stacks (**n**) and $(L_{av}/Dk)_{app}]$ versus $\mathbf{n}L_{av}$, respectively. By subtracting the two intercepts of these straight lines, we calculated the resistance of the two sets of boundary layers $L_{sl}/D_0k_0$ and $L_{bl}/D_0k_0$. The estimated thicknesses of the two sets of boundary layers [one set consists of the two layers: one between the lenses and the electrode and the other on top of the uppermost lens of each stacks ($L_{bl}$ ), and the second set that consists of all the layers sandwiched between the lenses in the stacks ($L_{sl}$)] were obtained from the boundary-layers' resistance, $L_{sl}/D_0k_0$ and $L_{bl}/D_0k_0$, and the permeability coefficient of water, $D_0k_0$ = 120 barrer.[31] From the "true" permeability coefficients and the measured harmonic mean thickness of the lenses, we calculated their "true" transmissibility coefficients (Table III).

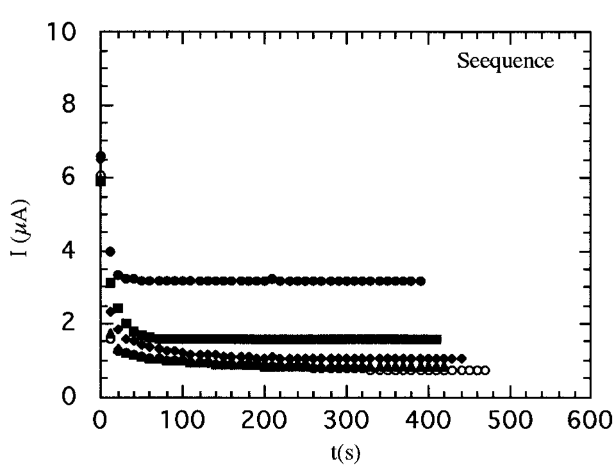


**Figure 3** Time course of the electric current obtained from (●) one, (■) two, (♦) three, (▲) four, and (○) five stacks for the Seequence lenses on the oxygen electrode.

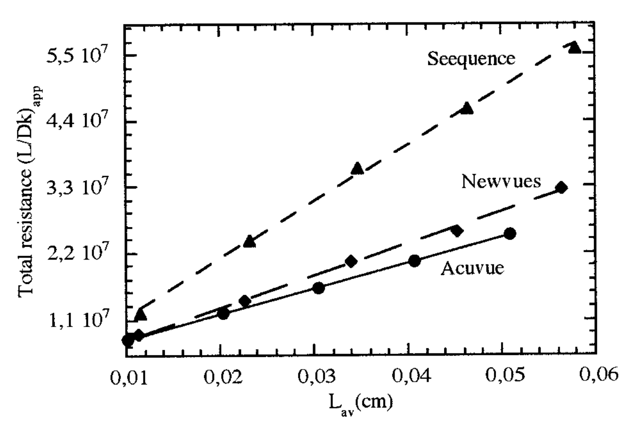


**Figure 5** Reciprocal of AOT versus the harmonic thickness (in cm) of stacks of one to five of Acuvue, Seequence, and Newvue contact lenses.

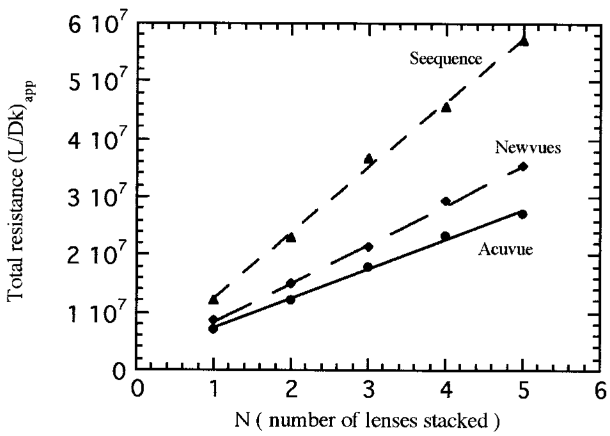


**Figure 6** Reciprocal of AOT versus the versus number of stacks for the three lenses studied.

The true permeability coefficients should be independent of the thickness of the lenses and of the method of determination. However, as seen in Table III, this is not always the case. Our values are closer to the coefficients determined by Weissman et al.[21,22] than of those reported by the manufacturers of the lenses. The agreement between the permeability coefficients determined by us and those determined by others is closer for the low hydration Seequence lenses than for the high hydration Newvue and Acuvue lenses. However, our *Dk* values are higher than those reported by Weissman et al. and by the manufacturers, except that the manufacturer reported the highest *Dk* for the Acuvue lenses. The large differences among the oxygen transmissibility coefficients reported by us and by previous investigators and by the manufacturers of the lenses reflect the different thicknesses, real or measured, that were used to obtain the transmissibility of the lenses.

The higher *Dk* of Newvue and Acuvue lenses obtained by us with eq. (4) differ more from those obtained by Weissman et al. with eq. (3) than does the *Dk* of the lower-hydration Seequence lenses. Equation (4) takes into account the resistance of all the sandwiched layers between the stacked lenses, while eq. (3) neglects these resistances. In our opinion, for the low permeability lenses, such as Seequence, the differences between the *Dk* obtained with eq. (3) or (4) are negligible because the resistance to the diffusion of oxygen by the fluid sandwiched layers between the stacked lenses is very small compared to the resistance of the lenses. On the other hand, the influence of the sandwiched-layers' resistance on the *Dk* increases with the oxygen permeability of the lenses. Therefore, although eq. (3) could be adequate to obtain the oxygen permeability coefficients of lower permeability materials, eq. (4) is required to obtain the true oxygen permeability coefficients of the high permeability materials.

In conclusion, eq. (3) is good for the calculation of the oxygen permeability coefficients of low-permeability (low hydration) standard hydrogel lenses, but we recommend the use of eq. (4) for the determination of the oxygen permeability coefficient of the standard hydrogel lenses of high water content (>50% water) and for the new generation of high gas permeable silicone-hydrogel lenses.[32] Similarly, eq. (6) [where $(L/Dk)_{\text{paper}}$ is the resistance of the moistened cigarette papers] should be used when stacks of hard oxygen permeable contact lenses with cigarette papers soaked in an electrolyte solution sandwiched between the individual lenses are used for the determination of the oxygen permeability of these hydrophobic lenses[11–13]:

$$\left(\frac{L}{Dk}\right)_{\text{app}} = \mathbf{n}\left[\left(\frac{L}{Dk}\right)_{\text{paper}} + \left(\frac{L_{\text{sl}}}{D_0k_0}\right)\right] + \left[\left(\frac{L_{\text{av}}}{Dk}\right)_{\text{lens}} + \left(\frac{L_{\text{bl}}}{D_0k_0}\right)\right] \quad (6)$$

**Table II AOTs $(Dk/L_{av})_{app}$, Permeability $(Dk)_{app}$, and Diffusion Coefficients of the Hydrogel Contact Lenses and Apparent Thickness of the Boundary Layers ($L_{bl}$) Between the Lenses and the Electrode and on the Surface of the Uppermost Lens and of the Sandwiched Layers ($L_{sl}$) Between the Stacked Lenses**

| Lens | $(Dk/L_{av})_{app}$ (barrer/cm) | $(Dk)_{app}$ (barrer) | $D_{app}$ ($10^{-7}$ cm²/s) | $L_{bl}$ (μm) | $L_{sl}$ (μm) |
|---|---|---|---|---|---|
| Seequence | 8.9 ± 0.5 | 10.3 ± 0.4 | 4.9±0.4 | 145 | 18 |
| Newvue | 14.7 ± 0.4 | 16.6 ± 0.4 | 8.1±0.7 | 209 | 22 |
| Acuvue | 19.5 ± 0.5 | 20.0 ± 0.4 | 3.6±0.3 | 284 | 27 |

**Table III Oxygen Permeability (barrer) and Transmissibility (barrer/cm) Coefficients of Hydrogel Contact Lenses**

| | This Article | | Weissman et al.[a 21,22] | | Manufacturers | |
|---|---|---|---|---|---|---|
| Lens | $(Dk)_{true}$ | $(Dk/L_{av})_{true}$ | $Dk$ | $Dk/L$ | $Dk$ | $Dk/L$ |
| Seequence | 10.5 ± 0.4 | 9.1 ± 0.5 | 9.0 | 15.0 | 8.4[a] | 27.0[a] |
| Newvue | 19.1 ± 0.4 | 16.9 ± 1.0 | 15.0 | 13.0 | 16.0 | 26.7 |
| Acuvue | 23.6 ± 0.4 | 23.1 ± 0.9 | 18.0 | 15.0 | 28.0 | 40.0 |

[a] Lens parameters: radius curvature, 8.4 mm (Seequence and Newvue), 8.8 mm (Acuvue); diameter, 14 mm; optical power −2.50 D.
[b] Measurement performed at 21°C for a lens with optical power −3.00 D.

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