Polymer-interaction driven diffusion of eyeshadow in soft contact lenses[☆]

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A B S T R A C T

Soft contact lenses used for the correction of ametropia are often made of hydrogel and silicone-hydrogel materials. Since they are placed directly on the surface of the eye and they are hydrated by tears, eye cosmetics can compromise the lens performance and, even worse, can be transported from an external environment to the ocular surface through the contact lens.

The diffusion of the dye component of a purple eyeshadow in soft contact lenses of different materials is here evaluated. Diffusivity is found to be typically higher in silicone-hydrogels than in hydrogels. In hydrogels, diffusivity is greater in the case of lower oxygen transmissibility. Despite differences between materials, absorbed mass of dye is much larger (10–100 times) than the expected mass by simple hydration and swelling of the contact lens. The most contaminated materials are also resistant to cleaning solutions. The results indicate that, notwithstanding the complexity of contact lens networks, diffusion of dye is found to follow Fick's law and it is driven by polymer-dye interaction, which governs lens hydration and swelling.

1. Introduction

Eye cosmetics typically contain pigments, waxes, oils, silicone components, and preservatives. Commercial cosmetics undergo safety testing for human use, but specific ocular changes associated with eye cosmetic are reported in the literature. For example, a very recent review summarizes current knowledge regarding the impact of cosmetic products on the eye, ocular surface, and tear film [1]. Stinging and burning, allergic conjunctivitis, allergic and irritant contact dermatitis, ocular infections, conjunctival pigmentation due to mascara and eyeliner, and eyeshadow mimicking orbital calcification were reported many years ago [2]. Conjunctival pigmentation secondary to eyelid cosmetics was also studied by light and electron microscopy [3]. The deposited materials were found to be ferritin particles and metal oxides [3]. Adverse reactions of cosmetics within the ocular surface were mainly attributed to the preservative benzalkonium chloride [4]. Extreme cases are those of ulcers associated with the use of mascara contaminated with *Pseudomonas aeruginosa* [5]. Makeup-human meibum interactions were recently studied in-vitro using infrared spectroscopy [6]. A makeup product was found to increase the lipid phase transition temperature when combined with human meibum

causing an increase in the order of the meibum-lipid hydrocarbon chains, which could have adverse effects on tear film stability. In general, practitioners are familiar with observing cosmetic residuals in tear fluid when they observe the eye under slit-lamp biomicroscope.

The use of both eye cosmetics and contact lenses (CLs) is also very frequent and deserves to be investigated for possible adverse events. Indeed, the impact of CL contamination by eye make-up may compromise the performance and physical properties of the CL and, even worse, transport of make-up from an external environment to the ocular surface through the CL is of clinical relevance because it can anticipate adverse interactions occurring at the ocular surface. Contamination of CLs can occur through the contaminated tear fluid and through the contact with hands. Contamination in the conjunctival sac by cosmetic and cleaning products was reported [7]. Pencil eyeliner was found to migrate most readily and to maximally contaminate the tear film when applied posterior to the lash line with possible implications for CL wearers [8]. In addition to eye contamination, deformation and swelling of the CLs were also observed for silicone-hydrogel materials without plasma polymerization coating when applying cosmetics and cleansing oil together or cleansing oil alone. In another study, the cleaning efficacy was also investigated for some brands of daily cleaners

for silicone hydrogel CLs contaminated by pencil eyeliner and cosmetic cleansing products [9]. Both hydrogel and silicone-hydrogel CLs in wet and dry conditions were recently investigated [10]. Hydrogels were found to resist cosmetic cleansing oil, while silicone-hydrogels have different degrees of resistance depending on the lens material. Modifications of silicone-hydrogel CL shape (diameter, sagittal depth, base curve) and optical power after in-vitro CL coating with cosmetics were recently reported [11]. The same group [12] also reported modifications of silicone-hydrogel CL surface by analyzing microscope images and wettability data. In this framework, many aspects related to interaction between CL materials, cosmetics, and tear film need additional investigation.

The aim of this work is to quantify and compare absorption of the dye component of a purple eyeshadow in soft CLs of different materials, with special focus on the diffusion process underlying the mass transfer phenomenon. Through the short-time solution of Fick's second law, dye diffusion coefficients (diffusivity) are deduced. Different CL materials, both hydrogels and silicone-hydrogels, are compared in terms of diffusion coefficient and efficacy of a multipurpose solution (MPS) in removing the dye component of the cosmetic, thus obtaining information on the polymer-cosmetic interaction.

2. Materials and methods

Different CL materials (-3.00 D) were investigated, as reported in Table 1, where t is the central CL thickness (provided by manufacturers), Dk/t is the CL oxygen transmissibility (provided by manufacturers), EWC is its equilibrium water content defined as

$$EWC = \frac{W_{hydr} - W_{dry}}{W_{hydr}}, \qquad (1)$$

W_{hydr} being the hydrated.CL weight and W_{dry} being the dehydrated-CL weight. EWC data reported in Table 1 are provided by manufacturers. Within an experimental error of 2%, the same values were deduced by measuring W_{hydr} and W_{dry} by a micro-balance Gibertini E42 (Italy) and applying Eq. (1).

The cosmetic is a purple highly concentrated loose powder eyeshadow (158, 40621, Kiko, Italy) made of talc, mica, paraffinum liquidum, magnesium stearate, lanolin oil, caprylic/capric triglyceride, phenoxyethanol, sodium dehydroacetate, diethyl-hexyl-syringylidenemalonate, and manganese violet dye (color index 77742), which are the typical components of eyeshadows (manganese dye is the typical colored component of purple ones).

The CLs were exposed to the cosmetic dissolved in 0.9% NaCl solution (Bausch & Lomb IOM saline solution) with cosmetic concentration equal to 1 mg/mL. Two procedures for evaluation of the relative mass of dye absorbed by a CL were applied. The first method is indirect because optical analyses are performed on solutions to obtain information on the absorbed mass in a CL. For the spectroscopic measurements of solutions, fused silica cuvettes were used with optical path of 1 mm. Absorbance measurements were performed between 900 and 220 nm by using a Jasco V-650 spectrophotometer (absorbance is
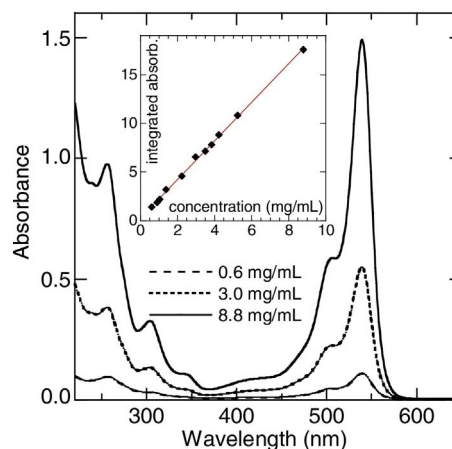

Fig. 1. Cosmetic absorbance spectra. Absorbance spectra of the cosmetic in 0.9% NaCl solutions with concentrations 0.6, 3.0, and 8.8 mg/mL. Inset: integrated absorbance from 540 to 610 nm as a function of cosmetic concentration in 0.9% NaCl solution. The line represents the result of linear regression of data (R = 0.999).

defined as $-\log_{10}$(transmittance)). The method was applied after verifying the linear dependence of optical absorbance on eyeshadow concentration. This was preliminary performed by measuring the optical absorbance of the cosmetic in 0.9% NaCl solution (Bausch & Lomb IOM saline solution) in the range of eyeshadow concentrations from 1.0 to 8.8 mg/mL. Spectra measured on different solutions are reported in Fig. 1. The integrated absorbance from 540 to 610 nm was calculated and is reported in the inset as a function of cosmetic concentration. Linear regression of data (R = 0.999) shows the linear dependence of absorbance on eyeshadow concentration. Even if the fraction of dye is not known, a linear dependence can be inferred also as a function of dye concentration, the dye being the colored component and representing a fixed fraction. The method consists in (i) measuring the optical absorbance of the cosmetic solution before and after CL exposure to 5 mL of solution in a glass vial and (ii) calculating the variation of dye concentration in solution after CL exposure compared to the dye concentration before exposure based on the measured values of absorbance. Indeed, the ratio between the two values of absorbance is equal to the ratio between the two dye concentrations due to their linear relationship. Therefore, the variation of optical absorbance of the solution is an indicator of dye absorption in CLs ranging from 0% to 100% (when the solution becomes transparent after the CL exposure). The relative mass of absorbed dye is defined as M_τ/M_{tot}, where M_τ is the mass of dye absorbed in one CL and M_{tot} is the total amount of dye in a vial (the dye fraction in 5 mg of eyeshadow, i.e. the mass of dye available in the vial for one CL). Analyses were repeated at least three times for each material. Since glass vials were used, the decrease of dye concentration due to possible dye adhesion to walls was also preliminary investigated. Experiments were carried out by filling vials with 5 mL of solution without any CL. After 24 h, the loss of dye was found to be $(5 \pm 2)\%$.

Table 1
Properties of the CLs investigated in this work.

| MATERIAL	FDA group	Brand	t (mm)	EWC (%)	oxygen Dk/t (Fatt units)
hilafilcon B	Hydrogel: II	Bausch & Lomb Soflens Daily	0.090	59	22
nelfilcon A	Hydrogel: II	Alcon Dailies Aquacomfort Plus	0.100	69	26
omafilcon A	Hydrogel: II	CooperVision Proclear 1 day	0.090	59	33
etafilcon A	Hydrogel: IV	Johnson & Johnson 1-day Acuvue Moist	0.084	58	33.3
filcon IV	Hydrogel: IV	Safilens Fusion 1 day	0.050	58	40
narafilcon A	Silicone-hydrogel: I	Johnson & Joh 1-day Acuvue TruEye	0.085	48	118
enfilcon A	Silicone-hydrogel: I	CooperVision Avaira	0.080	46	125
comfilcon A	Silicone-hydrogel: I	CooperVision Biofinity	0.080	48	160
filcon V	Silicone-hydrogel: n.a.	Safilens Open30	0.090	45	65
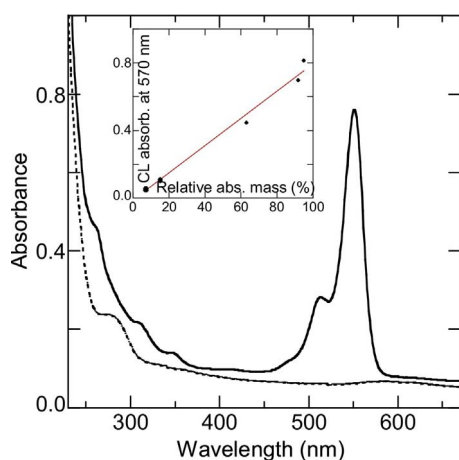
Fig. 2. Contact lens absorbance spectra. Absorbance spectra of nelfilcon A contact lens before (dotted line) and after (continuous line) exposure to the cosmetic solution (5 mL, 1 mg/mL) for 24 h. Inset: contact lens absorbance at 570 nm measured by the second method on different contaminated contact lenses compared to the corresponding relative mass of cosmetic absorbed per contact lens obtained by the indirect method. The line represents the result of linear regression of the data (R = 0.996).

The second method is direct because the optical analyses were performed directly on the CL. Also in this case, analyses were repeated at least three times for each material. Absorbance was measured by using the same Jasco V-650 spectrophotometer in the central portion (diameter 4 mm) of hydrated CLs mounted on a plastic support. The method consists in (i) measuring the absorbance spectra of contaminated and new CLs, (ii) calculating their difference at a specific wavelength λ (here λ = 570 nm), and, from this value, (iii) calculating the relative mass of dye in the CL (through a calibration curve). For example, Fig. 2 shows the absorbance spectra of a nelfilcon A CL before and after exposure for 24 h to the cosmetic solution (volume 5 mL, eyeshadow concentration 1 mg/mL). The calibration was obtained by comparing the absorbance at 570 nm measured on contaminated CLs (of different materials) with the relative absorbed mass of dye in the same CL obtained by applying the first indirect method on the solution. The inset of Fig. 2 shows these data and the line obtained by their linear regression (R = 0.996). It represents a calibration of the second method to obtain the relative mass of dye in a CL by the absorbance measured at 570 nm directly on the CL.

The second method was also used to verify the efficacy of a MPS (Safilens Open Reload) to remove cosmetic. Also in this case, analyses were repeated at least three times for each material. The procedure was the following: (i) CLs were removed from the cosmetic solution, (ii) they were rinsed with NaCl solution (Bausch & Lomb IOM solution) for few seconds, (iii) their absorbance at 570 nm was measured following the second method, (iv) they were exposed to the MPS (5 mL per CL), and (v) the CL absorbance at 570 nm was measured every 30 min for 6 h.

Fluorescence confocal micrographs were obtained using a Leica SP2 confocal laser scanning fluorescence microscope (Leica Microsystems, Germany). For each sample, a Z-stack was acquired to evaluate the cosmetic penetration depth. Excitation wavelength was 561 nm, where the eyeshadow is known to absorb the excitation incident light (Fig. 1). Fluorescence micrographs were taken by collecting the fluorescence of the sample in the spectral range between 570 nm and 650 nm.

3. Results

Fig. 3a shows the log–log plot of the relative absorbed mass M_τ/M_{tot} of dye in a nelfilcon A CL measured by the direct method as a function of the time τ of exposure to the cosmetic solution until 3 h of exposure. Similar results were obtained by the indirect method (here omitted). Similar log–log plots were obtained for all materials. The linear fitting
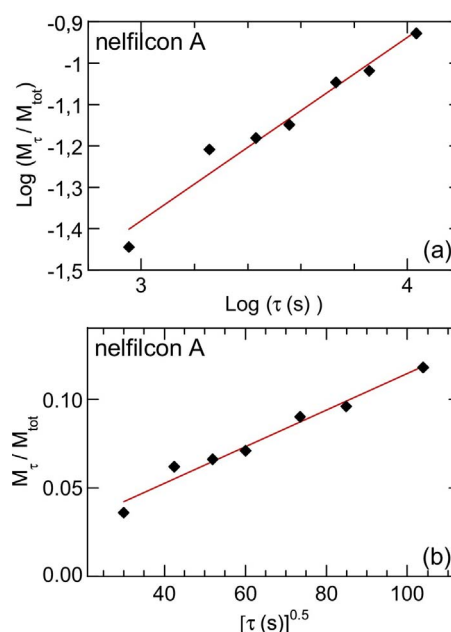

Fig. 3. Fick's diffusion. Relative mass M_τ/M_{tot} of dye absorbed in a contact lens measured by the direct method as a function of the time τ of exposure to the cosmetic solution (5 mL, eyeshadow concentration 1 mg/mL). The lines indicate the results of linear regression of the data ((a) R = 0.982, angular coefficient 0.444; (b) R = 0.982, angular coefficient 0.00103).

of the experimental data allowed to obtain the slopes for all the investigated CLs. All slopes were found to be compatible with 0.5 within an experimental error of 0.15.

Fig. 3b shows the same data as in Fig. 3a reported as relative mass M_τ/M_{tot} as a function of $\sqrt{\tau}$. The data linear regression allowed to deduce the angular coefficient, which is assumed to be equal to $\sqrt{\frac{16D_c}{t^2}}$ (as discussed in the following section), where D_c is the dye diffusion coefficient (diffusivity) in the CL and t is the CL thickness. Similar plots in the same time interval (τ ≤ 3 h) for all investigated materials allowed to deduce the different angular coefficients $\sqrt{\frac{16D_c}{t^2}}$, the ratio $\frac{D_c}{t^2}$, and finally D_c, by considering the known central thickness t of CL. The results are reported in Table 2.

Fig. 4 shows the relationship between measured dye diffusivity of the hydrogel materials and the reciprocal of their oxygen transmissibility. The line indicates the results of linear regression of the data (R = 0.914). For the investigated hydrogels, D_c and Dk/t are inversely proportional, whereas for silicone-hydrogels (inset of Fig. 4) there is no evidence of a relationship between D_c and Dk/t.

Analyses were also performed after a longer exposure to the cosmetic solution. Fig. 5 shows the relative absorbed mass M_τ/M_{tot} after

Table 2
Mean ± std dev for the different materials of (i) the angular coefficients $\sqrt{\frac{16D_c}{t^2}}$ (units s$^{-1/2}$) obtained by linear regression of $\frac{M}{M_{tot}}$ as a function of $\sqrt{\tau}$ for τ ≤ 3 h (see equation (2) in the text), (ii) the ratio $\frac{D_c}{t^2}$ (units 10^{-6} s^{-1}) between the diffusion coefficients D_c of the cosmetic in the CL and the square of the CL thickness t, and (iii) the diffusion coefficients (diffusivity) D_c (units 10^{-8} mm^2 s^{-1}).

| MATERIAL	$\sqrt{\frac{16D_c}{t^2}}$	$\frac{D_c}{t^2}$	D_c
hilafilcon B	0.00174 ± 0.00024	0.60 ± 0.16	0.48 ± 0.13
nelfilcon A	0.00094 ± 0.00018	0.173 ± 0.066	0.173 ± 0.066
omafilcon A	0.00082 ± 0.00003	0.1320 ± 0.0097	0.1069 ± 0.0078
etafilcon A	0.00113 ± 0.00037	0.25 ± 0.17	0.18 ± 0.12
filcon IV	0.00063 ± 0.00022	0.077 ± 0.055	0.019 ± 0.014
narafilcon A	0.00146 ± 0.00066	0.42 ± 0.38	0.30 ± 0.27
enfilcon A	0.00456 ± 0.00067	4.1 ± 1.2	2.61 ± 0.77
comfilcon A	0.00341 ± 0.00027	2.29 ± 0.36	1.46 ± 0.23
filcon V	0.00270 ± 0.00087	1.43 ± 0.92	1.16 ± 0.74
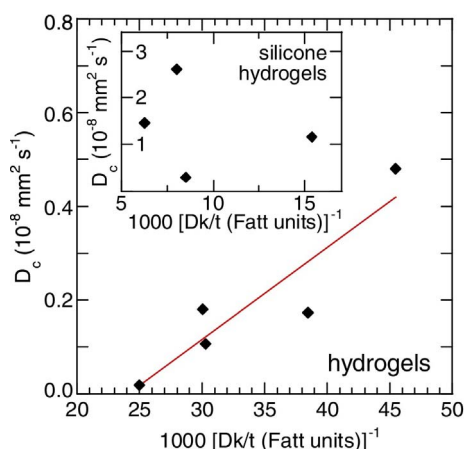
Fig. 4. D$_c$ vs Dk/t for hydrogels. Measured dye diffusivity D$_c$ of the hydrogel materials under investigation as a function of the reciprocal of their oxygen transmissibility Dk/t. The line indicates the results of linear regression of data (R = 0.914). Inset: Measured dye diffusivity D$_c$ of the silicone-hydrogel materials under investigation as a function of their oxygen transmissibility Dk/t.
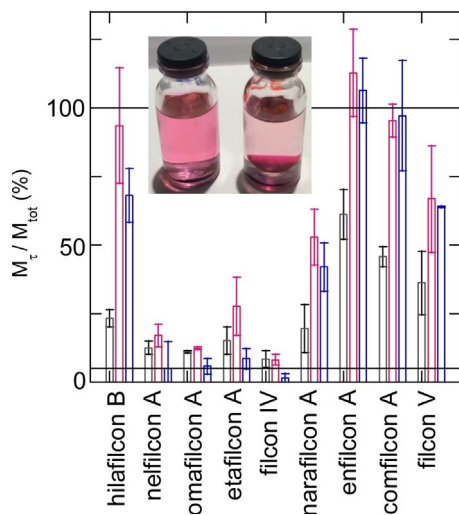

Fig. 5. Mass of dye absorbed by a contact lens. Relative mass of dye (mean on at least three independent analyses) absorbed in the different CLs obtained by the direct method after 5 h and 24 h of exposure to the cosmetic solution (first two columns) and after 6 h of further exposure to multipurpose solution (third column). The error bars indicate the std dev of the measured data. Horizontal lines indicate the upper experimental limit (100%, i.e. total mass of dye per CL available in the vial) and the lower limit (5%) corresponding to the relative mass of dye attached to the glass walls of the vial after 24 h as obtained by preliminary experiments. Inset: vial containing the cosmetic solution after few minutes (left) and after 24 h (right) of exposure of a hilafilcon B contact lens.

5 h and after 24 h of exposure (first two columns), when the maximum value is always reached (plateau). All silicone-hydrogels reached a relatively high contamination, reaching 100% for enfilcon A and comfilcon A. In our specific condition, 100% corresponds to the content of dye in a vial, thus representing the upper experimental limit for the absorbed mass in a CL, as indicated by the horizontal line in Fig. 5. Also one hydrogel material (hilafilcon B) out of five reached approximately the upper limit after 24 h. In the same figure, a lower limit (5%) is also indicated corresponding to the relative mass of dye attached after 24 h to the glass walls of the vial as obtained by preliminary experiments. The inset of Fig. 5 shows the solution after few minutes (left) and after 24 h (right) of exposure of a hilafilcon B CL.

In Fig. 5, a third column is reported for all materials showing MPS efficacy. As an example, the dye relative mass as a function of time during the CL exposure to MPS is reported for nelfilcon A in Fig. 6 (bottom). Similar analyses on all investigated materials allowed to
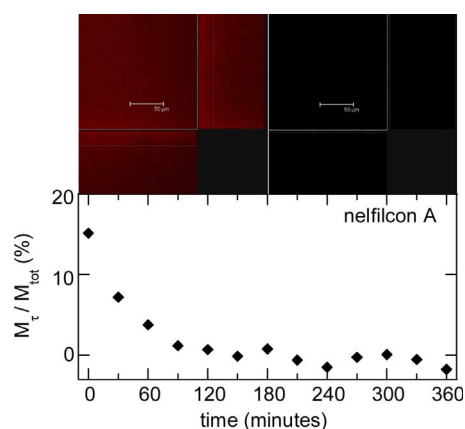

Fig. 6. Contact lens exposure to multipurpose solution. Relative mass of dye (bottom) in a nelfilcon A contact lens measured by the direct method as a function of time during the multipurpose solution exposure and fluorescence confocal micrographs (top) taken from the surface (main micrograph) and depth profiles along the grey lines (right and bottom of the main micrograph) for contaminated nelfilcon A contact lens (left) and nelfilcon A contact lens after 6 h of multipurpose solution exposure (right). The bars correspond to 10 μm.

obtain the values of the third column of Fig. 5, i.e. the relative mass of dye in a CL after MPS use. Also in this case, the behavior of silicone-hydrogels is different compared to hydrogels (excluding hilafilcon B). In the case of nelfilcon A, omafilcon A, etafilcon A, and filcon IV, dye was substantially removed from the CL. The quantitative data in Fig. 5 were obtained by optical absorption spectroscopy analyses (direct method). The cleaning efficacy can also be easily observed by collecting confocal microscopy micrographs of the CLs before and after the exposure to MPS by observing the fluorescence of the eyeshadow. For example, Fig. 6 (top) shows the fluorescence confocal micrographs for one of these materials. The micrograph of a contaminated nelfilcon A CL (left) shows homogeneous distribution of cosmetic in the CL, while no fluorescence is detected after 6 h of exposure to MPS (right). These micrographs are reported as visual examples. The two images were collected at the same experimental conditions, thus allowing an approximated luminance comparison. However, no detailed and extended fluorescence analyses were carried out because precise quantitative analyses are not trivial by fluorescence imaging. A completely different scenario was found for the other materials (third column of Fig. 5), the final mass being 80%-90% of the initial mass before MPS exposure.

4. Discussion

Log-log plots, such as that one reported in Fig. 3a, represent a method of verifying that the uptake follows normal Fickian behavior, if the slope is 0.5 [13]. From the experimental data, slopes were always found in the range between 0.35 and 0.65 for all investigated materials, in reasonable agreement with the prediction of Fick's diffusion theory [13]. Fick's laws describe the mass transport due to diffusion. In particular, first equation allows to predict the mass flux from a concentration gradient in space: a solute will move from a region of high concentration to a region of low concentration across a concentration gradient. Fick's second law predicts how diffusion causes the concentration to change with time. To model the uptake, Fick's second equation for one-dimensional, isothermal solute uptake in a polymer slab of thickness t was applied in the short-time approximation, namely for the first 3 h of exposure to the cosmetic solution. In this approximation, solution of Fick's equation is

$$\frac{M}{M_{tot}} = \sqrt{\frac{16 D_c}{t^2}}, \qquad (2)$$

which relates the fractional uptake of cosmetic, M$_\tau$/M$_{tot}$, to time τ. D$_c$ is the dye diffusion coefficient (diffusivity) in the CL. Given the above

boundary conditions, the angular coefficients $\sqrt{\frac{16D_c}{t^2}}$ were deduced from plots such as that one in Fig. 3b, together with the ratio $\frac{D_c}{t^2}$ and the diffusion coefficient D_c (Table 2). Three silicone-hydrogels out of four show a relatively high D_c, larger than $1 \cdot 10^{-8}$ mm^2 s^{-1}. On the contrary, hydrogels show at least one order of magnitude lower D_c than those for the silicone-hydrogels enfilcon A, comfilcon A, and filcon V. Due to the complexity of CL polymeric networks, several factors could account for this difference. Ionic surface charge can be excluded to explain the observed differences because the different FDA groups correspond to non ionic CLs and ionic CLs, but no substantial correlation was observed in this work between cosmetic contamination and FDA group. Interestingly, Fig. 4 shows the relationship between measured D_c of the hydrogel materials and reciprocal of oxygen transmissibility. For the investigated hydrogels, these two parameters D_c and Dk/t are found to be inversely proportional. It is also interesting that silicone-hydrogels do not show evidence of such a relationship between D_c and Dk/t.

For hydrogels, our experimental results indicate that large oxygen transmission is associated with low dye diffusivity. For these materials, higher oxygen transmission is known to be associated with larger EWC, i.e. larger mesh size (mesh size is the average distance between neighboring molecular chains). Therefore, our experimental results indicate that large mesh size in hydrogels is associated to low dye diffusivity. This suggests that uptake is driven by interaction of the dye with the hydrogel and that, for relatively large mesh size, the interaction between dye and polymer chains is minimized. In other words, diffusion of solute is not favored by large mesh (i.e. large EWC), as one might think, because the process is not driven by CL hydration and swelling, but it is driven by dye-polymer interaction. Silicone-hydrogels show a different behavior. Both oxygen transmissibility and dye diffusivity are relatively large compared to hydrogels, they are not correlated, and the dye-polymer interaction is found to be much stronger. All four silicone-hydrogels show similar EWC values, which are lower than EWC of the five hydrogels under investigation. This lower hydrophilicity corresponds to relatively low mesh size, which is expected to further favors the dye-polymer interaction.

A further confirmation that diffusion is driven by the interaction with the polymer is the large content of dye per CL. Indeed, despite differences between the materials, for all investigated CLs the dye content per CL is much larger than the expected absorbed mass by simple hydration of the CL. This is deduced by taking into consideration the water content of a CL (0.01–0.02 mL). This volume corresponds to 0.01–0.02 mg of eyeshadow (eyeshadow concentration 1 mg/mL) to be compared with the eyeshadow mass (5 mg) available in the vial. Although the dye component represents a fraction of the mass of eyeshadow, the expected relative absorbed mass of dye by simple hydration can be deduced as the ratio between 0.01–0.02 mg and 5 mg, i.e. less than 1%. The measured values are tens or hundreds times larger (Fig. 5).

The coefficients D_c were deduced by analyzing the dye uptake in short-time approximation. Also after a longer exposure, differences between hydrogels and silicone-hydrogels occurred and, for a few materials, the contamination reached the upper experimental limit (100%). In these cases, the strong absorption was also evident from the color of the solution after 24 h. For example, the inset of Fig. 5 shows the solution after few minutes (left) and after 24 h (right) of exposure of a hilafilcon B CL. The whole mass of dye available in the vial was absorbed by the CL, which became violet, making transparent the vial solution.

As far as the MPS efficacy is concerned, MPS was found effective in removing the dye from the less contaminated CLs. Also in this case, lower MPS efficacy can be attributed to the strong dye-polymer interaction occurring in silicone-hydrogels and in one of the investigated hydrogels.

5. Conclusions

Notwithstanding the complexity of CL polymeric networks, diffusion of the dye component of a cosmetic in soft CLs is found to follow Fick's behavior with different diffusivity for different investigated materials. Diffusion is found to be driven by dye-polymer interaction. This interaction is found to be stronger in silicone-hydrogels, thus explaining their higher dye diffusivity compared to hydrogels.

Among hydrogels, relatively high oxygen transmission (namely large EWC and mesh size) is found to be associated with low dye diffusion and vice versa. This aspect is interpreted as a consequence of reduced dye-polymer interaction in case of large mesh size. Therefore, diffusion of solute (dye) in hydrogel CLs is not favored in case of large EWC, as one might think, because solute diffusion is not driven by CL hydration and swelling, but it is driven by dye-polymer interaction.

Another consequence of the polymer-dye interaction is that, despite differences between the materials, for all investigated CLs the dye absorption is much larger (10–100 times) than the expected absorbed mass by simple CL hydration. Strong dye-polymer interaction is also found to reduce MPS efficacy.

Based on the present results on one specific cosmetic, silicone-hydrogels are found to be not recommended for prolonged wear for eyeshadow wearers. We highlight that this work is focused on the manganese dye component of the investigated eyeshadow. The dye absorption in CLs is not necessarily correlated to the absorption of other components, such as oils, mica, etc. Further investigations are required to study the interaction between these uncolored components and CL materials. Further analyses could also be performed on CLs worn by eyeshadow wearers to study the dye-polymer interaction in-vivo and possible effects on CL physical, optical, and clinical properties, both for the most contaminated materials and for the less contaminated ones. Based on the measured absorption spectrum of the dye, *in-vivo* observations could also be carried out under slit-lamp by illuminating the eye through a short-pass filter and by observing the eyeshadow fluorescence through a long-pass filter.

References

[1] A. Ng, K. Evans, R.V. North, L. Jones, C. Purslow, Impact of eye cosmetics on the eye, adnexa, and ocular surface, Eye Contact Lens 42 (2016) 211.
[2] F. Pascher, Adverse reactions to eye area cosmetics and their management, J. Soc. Cosmet. Chem. 33 (1982) 249.
[3] E.V. Platia, R.G. Michaels, W.R. Green, Eye cosmetic-induced conjunctival pigmentation, Ann. Ophthalmol. 10 (1978) 501.
[4] A. Malik, C. Claoué, Transport and interaction of cosmetic product material within the ocular surface: beauty and the beastly symptoms of toxic tears, Contact Lens Anterior Eye 35 (2012) 247.
[5] L.A. Wilson, D.G. Ahearn, Pseudomonas-induced corneal ulcers associated with contaminated eye mascaras, Am. J. Ophthalmol. 84 (1977) 112.
[6] M. Hunter, R. Bhola, M.C. Yappert, D. Borchman, D. Gerlach, Pilot study of the influence of eyeliner cosmetics on the molecular structure of human meibum, Ophthalmic Res. 53 (2015) 131.
[7] T. Tsukiyama, Y. Miyamoto, M. Fukuda, Y. Shimomura, J. Tsuchiya, H. Miura, Influence of eye cosmetics and cleansing products on contact lenses, J. Jap. Contact Lens Soc. 52 (2010) 101.
[8] A. Ng, K. Evans, V. North, C. Purslow, Migration of cosmetic products into the tear film, Eye Contact Lens 41 (2015) 304.
[9] J. Tsukiyama, Y. Miyamoto, M. Fukuda, Y. Shimomura, H. Miura, J. Tsuchiya, Contamination of silicone hydrogel contact lenses by cosmetics and the cleaning effect of contact lens daily cleaners, Invest. Ophthalmol. Vis. Sci. 52 (2011) 10.
[10] J. Tsukiyama, Y. Miyamoto, A. Kodama, M. Fukuda, Y. Shimomura, Cosmetic cleansing oil absorption by soft contact lenses in dry and wet conditions, Eye Contact Lens (2016), http://dx.doi.org/10.1097/ICL.0000000000000272 (in press).
[11] D. Luensmann, M. Yu, J. Yang, S. Srinivasan, L. Jones, Impact of cosmetics on the physical dimension and optical performance of silicone hydrogel contact lenses, Eye Contact Lens 41 (2015) 218.
[12] S. Srinivasan, H. Otchere, M. Yu, J. Yang, D. Luensmann, L. Jones, Impact of cosmetics on the surface properties of silicone hydrogel contact lenses, Eye Contact Lens 41 (2015) 228.
[13] P. Ritger, N. Peppas, A simple equation for description of solute release I. Fickian and non-Fickian release from non-swellable devices in the form of slabs, spheres, cylinders or discs, J. Controlled Release 5 (1987) 23.