Transmittance Calculations of a Sunscreen Film: Considerations of the Coating Geometry and its Probability Density Distribution

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ABSTRACT

Transmittance is an important parameter of sunscreen films such as sunscreen creams, coating materials, sunscreen sheets, etc. Even if the amounts of the sunscreen agent are the same, the transmittance greatly changes depending on the geometry of the coating interface. In this study, we calculate the transmittance considering the coating geometry and the probability density distribution of the thickness of the sunscreen film. We found analytical and numerical solutions of the transmittance in several model cases. It can be used for prediction of performance of the sunscreen film and for a fair comparative evaluation. Mathematical techniques in calculation of the transmittance are also explained in detail.

KEYWORDS: Integral equation theory; Inverse problem; Inverse analysis; Inverse function; Lebesgue integration; Cumulant expansion.
1. INTRODUCTION

Developments of sunscreen films such as sunscreen creams, coating materials, sunscreen sheets are important for a better life for us. In the developments, knowing a parameter of transmittance is one of the necessary things. The transmittance reveals the performance of the product. The parameter is also significant in developments of solar cells and photocatalysts. However, the transmittance largely depends on its coating geometry. Hence, it is important to understand relationship between the transmittance and the coating geometry. In this study, we calculate the transmittance considering the coating geometry and the probability density distribution of the thickness of the sunscreen film. We found analytical and numerical solutions of the transmittance in several model cases. It can be used for prediction of performance of the sunscreen film and for a fair comparative evaluation. In Chapter 2, the calculation theory of the transmittance is explained. In Chapter 3, the relationship between the transmittance and the coating geometry is shown. Probability density function (PDF) of thickness (height) of the coating film is also shown here. In Chapter 4, conclusion of this study and a few comments are written. In APPENDIX (Chapter 5), mathematical relationships derived in the process of the transmittance study are shown.

2. THEORY

2-1. Sunscreen film on a flat surface

To calculate the transmittance, we consider following coating film shown in Figure 1. In the model, the lower interface is completely flat, while the upper interface is modeled as a trigonometric function (sine or cosine function). The green solid line and $t_0$ both represent the average height of the film. The parameter $a$ is half amplitude of the trigonometric function and $t(x')$ represents the thickness as a function of
position $x'$. For geometric reason, range of $a$ is expressed as $0 \leq a \leq t_0$ ($-t_0 \leq a \leq t_0$ is also possible but we define $a$ as the value that is greater than or equal to 0 for explanatory simplicity). Note that the geometry should not follow the trigonometric function, but the height (thickness) must follow the arcsine distribution in the theory explained here. The model indicates that amounts of the coating materials are the same if each $t_0$ is the same. By the way, it has been reported that the thickness of the sunscreen cream on a biological skin accords with inverse gamma distribution [1][2]. Thus, Figure 1 is not the model of such a sunscreen cream, but that of an engineered film surface [3][4][5]. For simplicity, we use only a sine function as a trigonometric function.

**Figure 1.** Geometry of the film. The upper coating interface is expressed as a trigonometric function (sine or cosine function) and that of the lower is flat. The green solid line and $t_0$ both represent the average height of the film. The parameter $a$ is half amplitude of the trigonometric function and $t(x')$ represents the thickness as a function of position $x'$. In the theory, the geometry should not follow the trigonometric function, but the height (thickness) must follow the arcsine distribution.

As mentioned above, the geometry should not follow the trigonometric function, but the height (thickness) must follow the arcsine distribution. It means that the upper interface does not need to be a periodic shape. If we disregard reflection and scattering from the film, the transmittance ($T$) can be calculated by using following equation:
\[ T = \frac{1}{x} \int_{0}^{x} 10^{-\kappa t(x')} \, dx' = \frac{1}{x} \int_{0}^{x} e^{-\kappa t(x') \ln 10} \, dx', \quad (1) \]

where \( \kappa \) (m\(^{-1}\)) is extinction coefficient. The integration range is from 0 to \( x \), but from 0 to \( 2\pi n \) (\( n \) is a natural number) is also applicable. We are now considering the sufficiently long integration range \( (0 \ll x) \). The thickness function can be given by \( t(x') = t_0 + a \sin(cx') \). For simplicity, \( c \) is defined as a natural number, definition of which is not so unnatural because there are numerous humps within the beam cross section (within the integration range). Then, Eq. (1) is rewritten as

\[
T = \frac{1}{2\pi n} \int_{0}^{2\pi n} e^{-\kappa (\ln 10) [t_0 + a \sin(cx')]} \, dx' \\
= \frac{e^{-\kappa t_0 \ln 10}}{2\pi n} \int_{0}^{2\pi n} e^{-\kappa a (\ln 10) \sin(cx')} \, dx' \\
= \frac{10^{-\kappa t_0}}{2\pi} \int_{0}^{2\pi} e^{-\kappa a (\ln 10) \sin(cx')} \, dx'. \quad (2)
\]

It is likely that Eq. (2) is related to an integral form of the Bessel function of the first kind of 0th order \( (J_0) \):

\[
J_0(z) = \frac{1}{2\pi} \int_{-\pi}^{\pi} e^{iz \cos \theta} \, d\theta. \quad (3)
\]

\( J_0 \) can be rewritten as

\[
J_0(\text{bi}) = \frac{1}{2\pi} \int_{-\pi}^{\pi} e^{-b \cos \theta} \, d\theta = \frac{1}{2\pi} \int_{0}^{2\pi} e^{-b \sin \theta} \, d\theta. \quad (4)
\]

Hence, Eq. (2) is expressed as
\[ T = 10^{-\kappa t_0} J_0(ika \ln 10). \] (5)

An interesting thing is that the parameter \( c \) is disappeared in Eq. (5). That is, the transmittance does not depend on \( c \) (\( c \) was defined as a natural number in advance). Using Eq. (5), we calculate the maximum value of the transmittance (\( T_{\text{max}} \)). Substituting the maximum value of the half amplitude \( a = t_0 \), we obtain

\[ T_{\text{max}} = \lim_{a \to t_0} T(a) = 10^{-\kappa t_0} J_0(ika \ln 10). \] (6)

On the other hand, the minimum value of the transmittance (\( T_{\text{min}} \)) can be calculated when \( a = 0 \): \( T_{\text{min}} = 10^{-\kappa t_0} \). For instance, when \( \kappa = 1.7 \times 10^5 \text{ (m}^{-1}) \) and \( t_0 = 10^{-5} \text{ (m)} \), \( T_{\text{max}} / T_{\text{min}} \approx 10.5 \). Therefore, we can understand that geometrical control is important for control of the transmittance. It should be noted that this conclusion is not only for the sine function but also for a cosine function and a function that obeys the arcsine distribution.

2-2. Sunscreen film on a non-flat surface

We consider a following film model (see Figure 2). In the model, both of the interfaces are not flat but waving. The parameters \( a_1 \) and \( a_2 \) are half amplitudes of the trigonometric functions. The green solid lines indicate the average heights of the interfaces, the interval between them is \( t_0 \). In the theory, the geometry should not follow the trigonometric function, but each interfacial height must follow the arcsine distribution. In this case, the transmittance is expressed as

\[ T = \frac{1}{x} \int_0^x e^{-\kappa \ln 10} t(x') \, dx' = \frac{1}{x} \int_0^x e^{-\kappa \ln 10} \left[ t_1(x') - t_2(x') \right] \, dx'. \] (7)
Figure 2. Geometry of the coating interfaces. The upper and the lower coating interfaces are expressed as trigonometric functions (sine or cosine functions). The parameters $a_1$ and $a_2$ are half amplitudes of the trigonometric functions. The green solid lines indicate the average heights of the interfaces, the interval between them is $t_0$. In the theory, the geometry should not follow the trigonometric functions, but each interfacial height must follow the arcsine distribution.

where $t_1(x') = t_{01} + a_1 \sin(c_1 x')$ and $t_2(x') = -(t_0 - t_{01}) \mp a_2 \sin(c_2 x')$. The parameter ranges of $a_1$ and $a_2$ are $0 \leq a_1 \leq t_{01}$ and $0 \leq a_2 \leq t_0 - t_1$, respectively. Also in this case, we define $c_1$ and $c_2$ as natural numbers. It is not so unnatural because there are numerous humps within the beam cross section (within the integration range). To solve Eq. (7), we transform it as follows

$$T = \frac{1}{2\pi n} \int_0^{2\pi n} e^{-\kappa \ln(10) [t_{00} + a_1 \sin(c_1 x') \pm a_2 \sin(c_2 x')] } dx'$$

$$= e^{-\kappa t_{00} \ln(10)} \int_0^{2\pi n} e^{-\kappa a_1 \ln(10) \sin(c_1 x')} \cdot e^{-\kappa a_2 \ln(10) \sin(c_2 x')} dx'$$

$$= \frac{10^{-\kappa t_{00}}}{2\pi} \int_0^{2\pi} e^{-\kappa \ln(10) [a_1 \sin(c_1 x') \pm a_2 \sin(c_2 x')] } dx'. \quad (8)$$

However, it is considered that Eq. (8) cannot be solved analytically any further (it can be solved numerically). To obtain the analytical solution, we introduce a following limitation: $c_1 \gg c_2$. In this case, Figure 2 can be redrawn as Figure 3. The thickness distributions of them are the same when $c_1 \gg c_2$.
Figure 3. Geometrically transformed distribution of the thickness. The thickness distributions of Figures 2 and 3 are the same when $c_1 \gg c_2$. Colors of blue and red are related to those in Figure 2. Each length of the red line ($\Delta x'$) is sufficiently short. The blue curve is mounting on the red short line, height distribution of which obeys the arcsine distribution.

Since each length of the red line ($\Delta x'$) is sufficiently short and each blue curve mounting on the red line obeys the arcsine distribution, the partial transmittance at $x'$ in the range from $x'$ to $x' + \Delta x'$ is calculated as

$$T(x') = \frac{1}{\Delta x'} \int_{x'}^{x'+\Delta x'} 10^{-\kappa[t_0 \pm a_2 \sin(c_2x') + a_1 \sin(c_1x'')]\,dx''}$$

$$= \frac{1}{2\pi/c_1} \int_{x'}^{x'+2\pi/c_1} 10^{-\kappa[t_0 \pm a_2 \sin(c_2x') + a_1 \sin(c_1x'')]\,dx''}$$

$$= \frac{1}{2\pi/c_1} 10^{-\kappa[t_0 \pm a_2 \sin(c_2x')]} \int_{0}^{2\pi/c_1} 10^{-\kappa[a_1 \sin(c_1x'')]\,dx''}$$

$$= \frac{1}{2\pi} 10^{-\kappa[t_0 \pm a_2 \sin(c_2x')]} \int_{0}^{2\pi} 10^{-\kappa[a_1 \sin(c_1x'')]\,dx''}$$

$$= 10^{-\kappa[t_0 \pm a_2 \sin(c_2x')]J_0(i\kappa a_1 \ln 10)}.$$

(9)

Hence, the total transmittance is calculated as
\[ T = \frac{1}{2\pi n} \int_{0}^{2\pi n} T(x') \, dx' \]
\[ = \frac{1}{2\pi n} \int_{0}^{2\pi n} 10^{-\kappa [t_0 \pm a_2 \sin(c_2 x')] J_0(ik a_1 \ln 10) \, dx'} \]. \quad (10)

Furthermore, it can be rewritten as
\[ T = \frac{1}{2\pi n} \int_{0}^{2\pi n} e^{-\kappa (\ln 10) [t_0 \pm a_2 \sin(c_2 x')] J_0(ik a_1 \ln 10) \, dx'} \]
\[ = \frac{1}{2\pi} \int_{0}^{2\pi} e^{\mp \kappa a_2 (\ln 10) \sin(c_2 x')} J_0(ik a_1 \ln 10) \, dx' \]
\[ = 10^{-\kappa t_0} J_0(ik a_1 \ln 10) J_0(\pm ika_2 \ln 10). \] \quad (11)

Consequently, the analytical solution, the purpose of this section, is obtained. We note that “±” in \( J_0 \) can be omitted, because it is an even function. As shown in Eq. (11), the single integral in Eq. (7) was split into two integrals. In Chapter 5-1, we wrote general explanation of the split method.

By the way, we replace \( T \) in Eq. (11) with \( T_W \), and then we obtain
\[ T_W = 10^{-\kappa t_0} J_0(ik a_1 \ln 10) J_0(ik a_2 \ln 10). \] \quad (12)

In addition, we prepare a film that has the same half amplitude \( a_1 \) as \( T_W \) but \( a_2 \) is zero (the lower interface is flat). The transmittance of such a film \( (T_S) \) is given by
\[ T_S = 10^{-\kappa t_0} J_0(ik a_1 \ln 10). \] \quad (13)

As a consequence, ratio of \( T_W/T_S \) is calculated as
\[ T_W/T_S = J_0(ik a_2 \ln 10). \] \quad (14)
The ratio of $T_W/T_S$ can be used for prediction of the transmittance of a film that has the waves on both interfaces from the transmittance obtained with a film that has the waving and flat surfaces on respective interfaces, and vice versa. Moreover, if we use ratio of $T_{min}/T_W$, the transmittance of a film that has flat interfaces on both side can be predicted from the transmittance of a film that has the waves on both sides. These ratios are important for a fair comparative evaluation of the product performance.

Although a comment below is not so important for understanding of the transmittance property, we note that the way of thinking shown in Figure 3 and Eq. (12) can be applied to speed up of a numerical integration. If the integrand possesses a property shown in Figure 3, the numerical integration can be divided into two simple parts. The more explanation is written in Chapter 5-1.

2-3. PDF of the thickness

PDF of the thickness ($p(t)$) is important information in terms of the coating geometry. As the name implies, $p(t)$ takes following property:

$$\int_{t_{\min}}^{t_{\max}} p(t) dt = 1, \quad (15)$$

where $t_{\min}$ and $t_{\max}$ represent the minimum and the maximum values in $t$, respectively. The thickness takes positive values, and hence the integration range from 0 to $\infty$ is also applicable in Eq. (15). The transmittance can be also calculated by using $p(t)$, which is given below (the derivation process is explained in Chapter 5-2):
\[ T = \int_{t_{\text{min}}}^{t_{\text{max}}} e^{-\kappa \ln 10 \cdot t} p(t) dt. \] (16)

It is known that PDF of a sine function is an arcsine distribution. Thus, the transmittance of the film model shown in Figure 1 can be calculated by substituting the arcsine distribution into \( p(t) \), where \( t_{\text{min}} \) and \( t_{\text{max}} \) are \( t_0 - a \) and \( t_0 + a \), respectively.

There is the other calculation way of the transmittance, which is expressed by using an inverse function of cumulative PDF \( b(u) \) (the derivation process is explained in Chapter 5-2):

\[ T = \int_{0}^{1} e^{-\kappa \ln 10 \cdot b(u)} du. \] (17)

\( b(u) \) can be called the thickness as a function of the cumulative density function (CDF).

In summary, there are three ways to calculate the transmittance: Eq. (1), Eq. (16), and Eq. (17).

As mentioned above, the transmittance of the film model shown in Figure 1 can be calculated by substituting the arcsine distribution into \( p(t) \). However, can we obtain \( p(t) \) when the film model is Figure 2? The answer is yes. \( p(t) \) can be calculated by following a process below: (I) Numerically or analytically calculate the transmittance by using Eq. (7), the solution of which is a function of \( \kappa \ln 10 \); (II) View Eq. (16) as the Laplace transform,

\[ T = \int_{0}^{\infty} e^{-(\kappa \ln 10) \cdot t} p(t) dt, \] (18)

which can be realized due to the domain of definition and its range of \( p(t) \); (III) Perform the inverse Laplace transform numerically or analytically, and then \( p(t) \) can be obtained. In addition, we also explain acquisition process of PDF in the case of
\(c_1 \gg c_2\). In that case, the transmittance can be readily obtained from Eq. (12), which can be rewritten as

\[
T_W = 10^{-\kappa t_0} f_0(i\kappa a_1 \ln 10) f_0(i\kappa a_2 \ln 10)
\]

\[
= e^{-\kappa t_0 \ln 10} \frac{1}{2\pi} \int_0^{2\pi} e^{-\kappa a_1 \ln 10} \sin \theta \ d\theta \frac{1}{2\pi} \int_0^{2\pi} e^{-\kappa a_2 \ln 10} \sin \theta \ d\theta
\]

\[
= e^{-\kappa(a_1 + t_0 - a_1) \ln 10} \frac{1}{2\pi} \int_0^{2\pi} e^{-\kappa a_1 \ln 10} \sin \theta \ d\theta \frac{1}{2\pi} \int_0^{2\pi} e^{-\kappa a_2 \ln 10} \sin \theta \ d\theta
\]

\[
= \frac{1}{2\pi} \int_0^{2\pi} e^{-\kappa \ln 10} a_1 (1 + \sin \theta) \ d\theta \frac{1}{2\pi} \int_0^{2\pi} e^{-\kappa \ln 10} a_2 [(t_0 - a_1)/a_2 + \sin \theta] \ d\theta,
\]

where Eq. (4) is used for the derivation. Eq. (19) can be replaced using Eq. (16) as

\[
T_W = \frac{1}{2\pi} \int_0^{2\pi} e^{-\kappa \ln 10} a_1 (1 + \sin \theta) \ d\theta \frac{1}{2\pi} \int_0^{2\pi} e^{-\kappa \ln 10} a_2 [(t_0 - a_1)/a_2 + \sin \theta] \ d\theta
\]

\[
= \int_0^{2\pi} e^{-\kappa (\ln 10) t_0} p_1(t) \ dt \int_{t_0 - a_1 - a_2}^{t_0 + a_1 + a_2} e^{-\kappa (\ln 10) t} p_2(t) \ dt,
\]

where \(p_1(t)\) and \(p_2(t)\) are PDFs of heights of “\(a_1 (1 + \sin \theta)\)” and “\(a_2 [(t_0 - a_1)/a_2 + \sin \theta]\)”, respectively. The two integrals above can be seen as the Laplace transforms,

\[
T_W = \int_0^\infty e^{-\kappa (\ln 10) t} p_1(t) \ dt \int_0^\infty e^{-\kappa (\ln 10) t} p_2(t) \ dt.
\]

Mathematical form of Eq. (21) is related to the convolution integral in the Laplace transform. Hence, the inverse Laplace transform of \(T_W\) as a function of “\(\kappa \ln 10\)” is given by (related matter is discussed in Chapter 5-3)

\[
\frac{1}{2\pi i} \int_{-i\infty}^{+i\infty} e^{\kappa (\ln 10) t} T_W(\kappa \ln 10) d(\kappa \ln 10) = \int_0^t p_1(t') p_2(t - t') dt' = p_W(t).
\]
Since the solution of the inverse Laplace transform of the transmittance is PDF of the film thickness (see Eq. (18)), the integral on the right-hand side of Eq. (22) yields PDF in the case of Figure 3 \((p_W(t))\). From Eq. (22), a mathematical property that the convolution integral of two PDFs generates a new PDF is found. That is,

\[
\int_0^\infty \int_0^t p_1(t')p_2(t-t')dt'dt = \int_0^\infty p_W(t)dt = 1. \tag{22}
\]

The mathematical property above is one of the universal facts of PDF.

2-4. Thickness function with beat and its transmittance

Here, we challenge calculation of the transmittance in the case of \(c_1 \approx c_2\) \((c_1 \neq c_2)\) by applying the PDF integration. In Chapters 2-1, 2-2, and 2-3, we defined both \(c_1\) and \(c_2\) as natural numbers for simplicity and strictness. However, in this chapter, such restriction is eliminated. The elimination can be realized without problem, because the sufficiently long integration range has been introduced in the original equation (see corresponding sentence near blow Eq. (1)). When \(c_1 \approx c_2\) \((c_1 \neq c_2)\), sum of the sine functions makes beat. For instance, the shapes of “\(2\sin(2\pi x) + \sin(0.99 \times 2\pi x)\)” and “\(2\cos(2\pi x) + \cos(0.01\times2\pi x)\)” resemble each other in some degree where the former and the latter functions represent the cases of \(c_1 \approx c_2\) \((c_1 \neq c_2)\) and \(c_1 \gg c_2\), respectively. Especially, their PDFs of the film thicknesses are greatly similar in visual. Therefore, the transmittance in the case of \(c_1 \approx c_2\) \((c_1 \neq c_2)\) can be calculated by substituting PDF in the case of \(c_1 \gg c_2\) into Eq. (16). Since Eq. (16) connects to Eq. (7), the transmittance in the case of \(c_1 \approx c_2\) \((c_1 \neq c_2)\) can be approximately calculated by using Eq. (12) derived in the case of \(c_1 \gg c_2\). Using the PDF approximation, we were able to accurately calculate the transmittance up to 2 or 3 significant digits. The high accuracy
proves validity of the PDF approximation, and it means that approximate shape of PDF of beat can be calculated by using Eq. (22) of the convolution integral. This may be great advantage for other scientific calculations containing the beat functions. We note that an interesting point of the calculation is that the concept of the PDF integration (Eq. (16)) was inserted in the middle of the transmittance calculation.

2-5. Transmittances calculation with a pair of PDF models

Until now, we have used the sine function and its PDF (arcsine distribution) as a model of the coating film. In this chapter, we additionally introduce the inverse gamma distribution, the gamma distribution, the exponential distribution, the beta distribution, the chi-squared distribution, the Flory-Schulz distribution [6][7], etc. The inverse gamma distribution is known as a feasible model of the thickness distribution of the sunscreen cream on a biological skin [1][2]. The minimum values of the domains of the added distributions are all larger than or equal to 0. Although the other distributions may be not suitable model for the coating film thickness, they are introduced. This is because, the introductions are not mathematically problematic. If the lower interface is flat and PDF of the upper interface can be modeled as one of the PDFs mentioned above, the transmittance can be calculated by

\[ T = \int_{t_{\text{min}}}^{t_{\text{max}}} e^{-\kappa(\ln 10)t} p_j(t) dt = \int_{0}^{\infty} e^{-(\ln 10)t} p_j(t) dt, \quad (23) \]

where \( p_j(t) \) represents the selected PDF of the upper interface. On the other hand, when the lower interface is also not flat, the transmittance can be calculated by

\[ T = \int_{t_{\text{min}}}^{t_{\text{max}}} e^{-\kappa(\ln 10)t} p_{jk}(t) dt = \int_{0}^{\infty} e^{-(\ln 10)t} p_{jk}(t) dt, \quad (24) \]
\[ p_{jk}(t) = \int_0^t p_k(t')p_j(t - t')dt', \quad (25) \]

where \( p_k(t) \) and \( p_{jk}(t) \) represent the selected PDF of the lower interface and the mixed PDF. This calculation method can be utilized when geometrical relationship between the upper and the lower interfaces corresponds to the situation in Chapters 2-2 (Figure 3) or Chapter 2-4 (beat). It is convenient way to elucidate the transmittance property, because the calculation is relatively simple and there is no need to prepare a concrete geometrical model.

3. RESULTS AND DISCUSSION

In this chapter, we show results from the calculations briefly. Figure 4 shows the ratio of transmittance vs amplitude of the upper sine function. Figure 5 shows PDF of the film thickness inversely calculated from a film that has waving interfaces on both sides.

![Graph](image)

**Figure 4.** Ratio of transmittance \((T/T_{\text{min}})\) vs half amplitude of the upper sine function \(a_1\). Here, the half amplitude of the lower sine function \(a_2\) is 0 (thin red curve) or 5 μm (thick green curve). The
amounts of the sunscreen creams on a unit area are $10^{-3}$ cm$^3$/cm$^2$ (average thickness is 10 μm), which are the same in all of the conditions. The extinction coefficient $\kappa$ (m$^{-1}$) is set to 2×10$^5$ m$^{-1}$.

As a supplement, a numerically integrated result (black dotted curve) is also shown where the analytical solution of Eq. (12) is not used. The dotted curve was obtained from a numerical computation with the same condition as the thick green curve except for the parameters of $c_1$ and $c_2$. In the thick green curve, $c_1 \gg c_2$, while in the black dotted curve, $c_1/5 = c_2$.

**Figure 5.** $p_w(t)$ (red) calculated by the convolution integral, where the thickness function is $t(x') = 4 + \sin(c_1x') + 2\sin(c_2x')$ and the condition is $c_1 \gg c_2$. It is related to Figure 2 with the condition $c_1 \gg c_2$, leading to Figure 3. Arcsine distribution (painted area) from 2 to 6 is also shown as a reference, which is related to PDF of the film thickness in Figure 1. As explained in Chapter 2-4, the convolution integral generates approximate form of PDF of beat, also. Hence, one can see the red curve as the approximate form of PDF of beat.

As shown in Figure 4, the transmittance increases as the half amplitude increases. The result is caused by partially formed thin areas of the film. The dotted curve was obtained from a numerical computation with the same condition as the thick green curve except for the parameters of $c_1$ and $c_2$. In the thick green curve, $c_1 \gg c_2$, while in
the black dotted curve, $c_1/5 = c_2$. From the comparison among the thick green curve, black dotted curve, and the other numerical results (not shown), we found that if $c_1$ is about 5 times larger than $c_2$, the accuracy of the analytical solution (Eq. (12)) is quite high.

The blue curve shown in Figure 5 is called the arcsine distribution, which is one of famous PDFs. In contrast, the red curve in Figure 5 has an unfamiliar shape. However, the shape is reasonable, because its rough shape can be imagined from Figure 3. From Figure 5, it is found that the PDF of the film thickness in the case of the both upper and lower interfaces are waving decays like a mountain slope on both side and has peaks in the middle area. The shape will change depending on the substituted pair of PDFs.

4. CONCLUSION

In summary, we have shown calculation method of the transmittance by introducing several film models. It has been shown that the transmittance is increased as the roughness is increased. PDF of the thickness has been also shown by using the inverse calculation method. The transmittance is one of the important parameters for developments of sunscreen creams, coating materials, sunscreen sheets, solar cells, and photocatalysts. The achievements obtained in the present study are significant for a fair comparative evaluation. Several mathematical techniques have been explained, which may be useful for other scientific calculations. In the future, we will use the mathematical techniques to challenge and solve several inverse problems in physical chemistry and biophysics.
5. APPENDIX

Here, we show some mathematical relationships derived in the process of the transmittance study. These relationships may be useful in other scientific calculations.

5-1. Splitting of a single integral into two integrals

We consider a following integral \( F_0(s) \),

\[
F_0(s) = \frac{1}{x_2 - x_1} \int_{x_1}^{x_2} e^{-s[h_0 + h_1(x) + h_2(x)]} \mathrm{d}x, \tag{26}
\]

where \( s, h_0, h_1(x), \) and \( h_2(x) \) are a variable of \( F_0 \), a constant, arbitrary periodic functions 1 and 2, respectively. The integral is performed in following condition: \( 0 \leq x_1 \leq x_2 \). If length of one cycle of the periodic function 1 (\( \lambda_1 \)) is much shorter than that of the periodic function 2 (\( \lambda_2 \)), the integral can be seen as Figure 3. Then, we perform the integration from \( x \) to \( x + \Delta x \) before starting the integration from \( x_1 \) to \( x_2 \):

\[
F_0^*(s; x) = \frac{1}{\Delta x} \int_{x}^{x+\Delta x} e^{-s[h_0 + h_1(x) + h_2(x)]} \mathrm{d}x. \tag{27}
\]

Since \( \lambda_1 \) is sufficiently shorter than \( \lambda_2 \), Eq. (27) can be rewritten as

\[
F_0^*(s; x) = e^{-s[h_0 + h_2(x)]} \frac{1}{\Delta x} \int_{x}^{x+\Delta x} e^{-sh_1(x)} \mathrm{d}x. \tag{28}
\]

When \( \lambda_1 \) is sufficiently shorter than \( \Delta x \), \( \Delta x \) can be expressed as \( \Delta x = (n + \Delta n)\lambda_1 \), where \( n \) is a natural number and \( 0 \leq \Delta n \leq 1 \). Then Eq. (28) becomes

\[
F_0^*(s; x) = e^{-s[h_0 + h_2(x)]} \frac{1}{(n + \Delta n)\lambda_1} \int_{x}^{x+(n+\Delta n)\lambda_1} e^{-sh_1(x)} \mathrm{d}x
\]
where we used approximation that \( (n + \Delta n) = n \). This is because, \( \lambda_1 \) is sufficiently shorter than \( \Delta x \), corresponding to that \( n \) is sufficiently larger than \( \Delta n \). Using Eq. (29), Eq. (26) can be rewritten as

\[
F_0(s) = \frac{1}{x_2 - x_1} \int_{x_1}^{x_2} F_1^*(s; x) \, dx
= \frac{1}{x_2 - x_1} \int_{x_1}^{x_2} e^{-s[h_0+h_2(x)]} \frac{1}{\lambda_1} \int_0^{\lambda_1} e^{-sh_1(x')} \, dx' \, dx
= e^{-sh_0} \left( \frac{1}{x_2 - x_1} \int_{x_1}^{x_2} e^{-sh_2(x)} dx \right) \left( \frac{1}{\lambda_1} \int_0^{\lambda_1} e^{-sh_1(x')} \, dx' \right).
\] (30)

Therefore, the single integral can be split into two integrals when the situation of Figure 3 (\( \lambda_2 \gg \lambda_1 \)) is realized. In the present study, use of Eq. (30) enabled us to obtain the analytical solution of the transmittance (Eq. (11)). This mathematical technique is also applicable when sum of \( h_1(x) \) and \( h_2(x) \) generates beat (\( \lambda_2 \approx \lambda_1 \) and \( \lambda_2 \neq \lambda_1 \)) and the integration range is sufficiently long (see Chapter 2-4). After the derivation, we realized that concept of the mathematical technique is somewhat related to that of the Riemann-Lebesgue lemma.

5-2. Roles of PDF and an inverse function

Here, we introduce an integral equation \( (F_1(s)) \),

\[
F_1(s) = \frac{1}{x_2 - x_1} \int_{x_1}^{x_2} f(s, h(x)) \, dx,
\] (31)

where \( f \) and \( h \) are arbitrary functions. For example, the above equation can be seen as a
general form of $F_0(s)$. Replacing the integral variable $x$ by $h$, Eq. (31) can be rewritten as

$$F_1(s) = \frac{1}{x_2 - x_1} \int_{h(x_1)}^{h(x_2)} f(s, h) \frac{dx}{dh} dh. \quad (32)$$

We note that $dx/dh$ is a function of $h$ and it can be calculated from $1/(dh/dx)$. Here, we consider an inverse function of $h(x)$. If $h(x)$ is a one-to-one correspondence in the integral range from $x_1$ to $x_2$, its inverse function can be systematically obtained, which we express $v(h) = x$. Derivative of the inverse function is expressed as

$$\frac{dv}{dh} = \frac{dx}{dh}. \quad (33)$$

In addition, a following relationship holds:

$$\frac{1}{x_2 - x_1} \int_{h(x_1)}^{h(x_2)} \frac{dv}{dh} dh = \frac{1}{x_2 - x_1} \int_{v(h(x_1))}^{v(h(x_2))} dv = 1. \quad (34)$$

Hence, $(dx/dh)/(x_2 - x_1)$ written in Eq. (32) can be seen as PDF (probability density function): $p(h)$. Then, Eq. (32) is rewritten as

$$F_2(s) = \int_{h(x_1)}^{h(x_2)} f(s, h)p(h) dh, \quad (35)$$

where we defined the PDF integral above as $F_2(s)$ ($= F_1(s)$) to distinguish it from the original integral form (Eq. (31)). If $h(x)$ is not the one-to-one correspondence in the integral range, $F_2(s)$ can be expressed as
\[ F_2(s) = \int_{h(x_1)}^{h(x_{m_1})} f(s, h)p_{m_1}(h) \, dh + \int_{h(x_1)}^{h(x_{m_2})} f(s, h)p_{m_2}(h) \, dh + \cdots + \int_{h(x_1)}^{h(x_{m_N})} f(s, h)p_{mN+1}(h) \, dh, \quad (36) \]

where \( x_{m_j} \) \((j = 1, 2, \ldots, N)\) is a joint position between the one-to-one correspondence domains and \( p_{m_j}(h) \) is PDF of \( h \) within the corresponding integral range (\( p_{m_j}(h) \) is zero outside the corresponding integral range). It can be simplified as

\[ F_2(s) = \int_{h_{\text{min}}}^{h_{\text{max}}} f(s, h) \left( \sum_{j=1}^{N+1} p_{m_j}(h) \right) \, dh. \quad (37) \]

where \( h_{\text{min}} \) and \( h_{\text{max}} \) are the minimum and the maximum values of \( h(x) \) in the domain from \( x_1 \) to \( x_2 \). Hence, when \( h(x) \) is a periodic function and sum of \( p_{m_j}(h) \) (the whole PDF from \( x_1 \) to \( x_2 \)) is known, \( F_1(s) \) can be calculated from \( F_2(s) \) (Eq. (37)).

By the way, we define \( F_3(s) \) as follows:

\[ F_3(s) = \int_0^1 f(s, b(u)) \, du, \quad (38) \]

which has an analogous form compared with Eq. (31). In this stage, a concrete expression of \( b(u) \) is not known, but it will be fabricated so that a following equation holds: \( F_3(s) = F_2(s) = F_1(s) \). The concrete expression of \( b(u) \) can be derived below. The PDF integral form of Eq. (38) is

\[ \int_0^1 f(s, b(u)) \, du = \int_{h(0)}^{h(1)} f(s, h)p(h) \, dh. \quad (39) \]

Here, please recollect that \( p(h) \) was made from \( h(x) \) of the one-to-one correspondence domain through the three steps.
Step (a): Obtain the inverse function of $h(x)$ ($v(h) = x$).

Step (b): Differentiate $v(h)$ with respect to $h$.

Step (c): Divide the derivative of $v(h)$ by $x_2 - x_1$, which is equal to $p(h)$.

Using the same calculation steps (a)-(c), $p(h)$ can be obtained from $b(u)$. However, the concrete expression of $b(u)$ is not known and $p(h)$ is known instead in this stage. It is considered that the concrete expression of $b(u)$ can be obtained by tracing the inverse route of steps (a)-(c).

Step (C): Multiply $p(h)$ by 1 ($= x_2 - x_1$).

Step (B): Integrate $p(h)$ with respect to $h$, which corresponds to CDF.

Step (A): Obtain the inverse function of CDF, which is equals to $b(u)$.

Introducing arbitrary examples, we calculated $F_3(s)$ by substituting $b(u)$ obtained from steps (C)-(A), and then we confirmed that $F_3(s)$ exactly matches $F_2(s)$ and $F_1(s)$. It indicates that if the integrated curve (or line segment) of $h(x)$ takes the one-to-one correspondence and its gradient is always positive in the domain from $x_1$ to $x_2$, $h(x)$ can be also inversely calculated from $p(h)$ through steps (C)-(A). However, please do not forget that steps (C), (B), and (A) are replaced that “Multiply $p(h)$ by $(x_2 - x_1)$”, “Integrate $(x_2 - x_1)p(h)$ with respect to $h$, and then get the cumulative function”, and “Obtain the inverse function of the cumulative function being $B(u)$, and then $h(x)$ is generated as follows: $h(x) = B(x - x_1)$”, respectively.

After we derived the mathematical technique, we found that the concept of the integral technique in $F_2(s)$ is similar to that of the Lebesgue integration. In addition, we note that if one would like to calculate $F_1(s)$ with $f(s, h(x)) = \exp(-sh(x))$ but it is
analytically impossible, we recommend the researcher to use both \( p(h) \) \((F_2(s))\) and the cumulant expansion. It is one of the compromise schemes.

5-3. Inverse transforms of specified \( F_1(s) \) and \( F_3(s) \) generate PDF

We have explained above that \( F_1(s), F_2(s), \) and \( F_3(s) \) are equally connected. In this chapter, it is explained that the inverse Laplace and the inverse Fourier transforms of specified \( F_1(s) \) and \( F_3(s) \) generate PDF. In the integrand of \( F_2(s) \), there is \( p(h) \). Since \( p(h) \) is PDF, it is completely 0 outside the integration range. Hence, \( F_2(s) \) can be rewritten as

\[
F_2(s) = \int_0^\infty f(s,h)p(h) \, dh, \quad (40)
\]

or

\[
F_2(s) = \int_{-\infty}^\infty f(s,h)p(h) \, dh. \quad (41)
\]

If \( f(s,h) \) is \( \exp(-sh) \), Eq. (40) corresponds to the Laplace transform. Therefore, the inverse Laplace transform of \( F_1(s) \) and \( F_3(s) \) generates \( p(h) \) as follows:

\[
\frac{1}{2\pi i} \int_{\gamma-i\infty}^{\gamma+i\infty} \exp(sh) \left[ \frac{1}{x_2 - x_1} \int_{x_1}^{x_2} \exp(-sh(x)) \, dx \right] \, ds = p(h), \quad (42)
\]

where \( b(u) \) is the inverse function of CDF, i.e., the inverse function of cumulative PDF. It should be noted that \( h(x) \geq 0 \) in Eq. (42) \((x \) can takes both positive and negative
values), because $p(h) \geq 0$ and the integration range of the variable $h$ in Eq. (40) is from 0 to $\infty$. This restriction originates from relationship between layouts of CDF and its inverse function. For example, when $h(x) = 1 + \sin x$, the inverse Laplace transform generates its PDF (arcsine distribution):

$$
\frac{1}{2\pi i} \int_{y-i\infty}^{y+i\infty} \exp(sh) \left[ \frac{1}{2\pi} \int_0^{2\pi} \exp(-s(1 + \sin x)) \, dx \right] \, ds
= \frac{1}{\pi \sqrt{1 - (h - 1)^2}} H_1(h) H_1(2 - h),
$$

where $H_1$ is the Heaviside step function containing a property that $H_1(0) = 1$. Since the analytical solution of the function in the square bracket above is $J_0(is)e^{-s}$ ($J_0$ is the Bessel function of the first kind of 0th order, see Eq. (4)), the inverse Laplace transform of $J_0(is)e^{-s}$ is the arcsine distribution. Likewise,

$$
\frac{1}{2\pi i} \int_{y-i\infty}^{y+i\infty} \exp(sh) \left[ \frac{1}{2\pi m} \int_0^{2\pi m} \exp(-s(1 + \sin x)) \, dx \right] \, ds
= \frac{1}{\pi \sqrt{1 - (h - 1)^2}} H_1(h) H_1(2 - h),
$$

where $m$ is a natural number. When $m$ is infinitely large, we obtain (see the denominator and the integral range)

$$
\frac{1}{2\pi i} \int_{y-i\infty}^{y+i\infty} \exp(sh) \left[ \frac{1}{2\pi m + 1} \int_0^{2\pi m + 1} \exp(-s(1 + \sin x)) \, dx \right] \, ds
= \frac{1}{\pi \sqrt{1 - (h - 1)^2}} H_1(h) H_1(2 - h).
$$

On the other hand, when $m$ is not so large, it is calculated as
\[
\frac{1}{2\pi i} \int_{\gamma-i\infty}^{\gamma+i\infty} \exp(sh) \left[ \frac{1}{2\pi m + 1} \int_0^{2\pi m + 1} \exp(-s(1 + \sin x)) \, dx \right] ds \\
= \left( \frac{m}{\sqrt{1 - (h - 1)^2}} H_1(h) H_1(2 - h) + \frac{1}{\sqrt{1 - (h - 1)^2}} H_1(h - 1) H_1(1 + \sin(1) - h) \right) / (1 + m\pi), \tag{46}
\]

where Eq. (37) is also utilized to solve. As far as we know, an analytical solution of the integral in the square bracket above is not known, however, its analytical solution of the inverse Laplace transform can be readily obtained from our mathematical technique. It is an interesting point of the technique. An equation below may be helpful to understand Eq. (46).

\[
\frac{1}{2\pi i} \int_{\gamma-i\infty}^{\gamma+i\infty} \exp(sh) \left[ \int_0^1 \exp(-ss\sin x) \, dx \right] ds = \frac{1}{\sqrt{1 - h^2}} H_1(h) H_1(\sin(1) - h). \tag{47}
\]

This kind of the inverse calculation can be also performed in the case of the inverse Fourier transform. The inverse Fourier transform of \( F_1(s) \) and \( F_3(s) \) generates \( p(h) \) as follows:

\[
\frac{1}{2\pi} \int_{-\infty}^{\infty} \exp(ish) \left[ \frac{1}{x_2 - x_1} \int_{x_1}^{x_2} \exp(-ish(x)) \, dx \right] ds \\
= \frac{1}{2\pi} \int_{-\infty}^{\infty} \exp(ish) \left[ \int_0^1 \exp(-isb(u)) \, du \right] ds = p(h). \tag{48}
\]

We note that \( x \) and \( h(x) \) can take both positive and negative values. \( b(u) \) is the inverse function of CDF. This restriction originates from relationship between layouts of CDF and its inverse function. Since the integration range of the Fourier transform of \( p(h) \) is from \(-\infty\) to \( \infty \), \( h \) can takes a negative value also.
In studies of liquids, colloidal dispersions, small angle scatterings [8], the Fourier transform for a radial distribution function is often used. Using its forward and inverse transforms, a following mathematical relationship can be also written:

\[
(4\pi)^2 \int_{0}^{\infty} \frac{ssin(sh)}{h} \left[ \frac{1}{x_2 - x_1} \int_{x_1}^{x_2} \frac{h(x)sin(sh(x))}{s} dx \right] ds = \left(4\pi\right)^2 \int_{0}^{\infty} \frac{ssin(sh)}{h} \left[ \int_{0}^{1} \frac{b(u)sin(sb(u))}{s} du \right] ds = p(h). \tag{49}
\]

It should be noted that \( h(x) \geq 0 \) in Eq. (49) (\( x \) can takes both positive and negative values), because \( p(h) \geq 0 \) and the integration range of the variable \( h \) in Eq. (40) corresponding to the Fourier transform for the radial distribution function is from 0 to \( \infty \). This restriction originates from relationship between layouts of CDF and its inverse function.

Generally, an integral transform (forward version) can be expressed as

\[
F(s) = \int_{r_1}^{r_2} K_{for}(s, h)p(h) dh, \tag{50}
\]

where \( K_{for} \) represents the integral kernel of the forward transform. \( r_1 \) and \( r_2 \) follow an integral range of the forward transform. In addition, the corresponding inverse transform is written as

\[
p(h) = \int_{r_3}^{r_4} K_{inv}(s, h)F(s) ds, \tag{51}
\]

where \( K_{inv} \) represents the integral kernel of the inverse transform. \( r_3 \) and \( r_4 \) follow an integral range of the inverse transform. For example, Hankel, Mellin, Hartley, Kontorovich-Lebedev, and Laguerre transforms are also categorized as the integral
transform. Substituting the mathematical technique derived here into the general integral transform, a following relationship can be obtained:

$$\int_{r_3}^{r_4} K_{\text{inv}}(s,h) \left[ \frac{1}{x_2 - x_1} \int_{x_1}^{x_2} K_{\text{for}}(s,h(x)) \, dx \right] \, ds$$

$$= \int_{r_3}^{r_4} K_{\text{inv}}(s,h) \left[ \int_{0}^{1} K_{\text{for}}(s,b(u)) \, du \right] \, ds = p(h) \quad (52)$$

It should be noted that the value of $h(x)$ and the domain from $x_1$ to $x_2$ follows each rule of the integral transform and the layout relationship. Since the Heaviside step functions are multiplied by $p(h)$ implicitly, the right hand side of Eq. (52) can be written as $p(h)H_1(h - h_{\text{min}})H_1(h_{\text{max}} - h)$, where $h_{\text{min}}$ and $h_{\text{max}}$ are the minimum and the maximum values of $h(x)$ in the domain from $x_1$ to $x_2$. Eq. (52) is one of the great achievements in the present study. As a supplement, we show a following equation:

$$\int_{r_1}^{r_2} K_{\text{for}}(s,h) p(h) \, dh = \frac{1}{x_2 - x_1} \int_{x_1}^{x_2} K_{\text{for}}(s,h(x)) \, dx = \int_{0}^{1} K_{\text{for}}(s,b(u)) \, du. \quad (53)$$

The equation represents the forward transform of PDF.

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