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Supporting Information

Pigmentation of White, Brown, and Green Chicken Eggshells Analyzed by Reflectance, Transmittance, and Fluorescence Spectroscopy

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The Bi-Layer Model

The reflectance and transmittance of two separate layers i=1,2 are described with the KM-equations inserting a common scattering coefficient S, individual absorption coefficients K_1, K_2, and individual layer thicknesses d_1, d_2 as

R_i = (Exp(b_i * S * d_i) - Exp(-b_i * S * d_i)) /(a_i *(Exp(b_i * S * d_i) - Exp(-b_i * S * d_i)) + b_i * (Exp(b_i * S * d_i) + Exp(-b_i * S * d_i))

T_i = 2b_i /(a_i *(Exp(b_i * S * d_i) - Exp(-b_i * S * d_i)) + b_i * (Exp(b_i * S * d_i) + Exp(-b_i * S * d_i))

where $a_i = (K_i + S) / S$, and $b_i = Sqr(a_i^2 -1)$

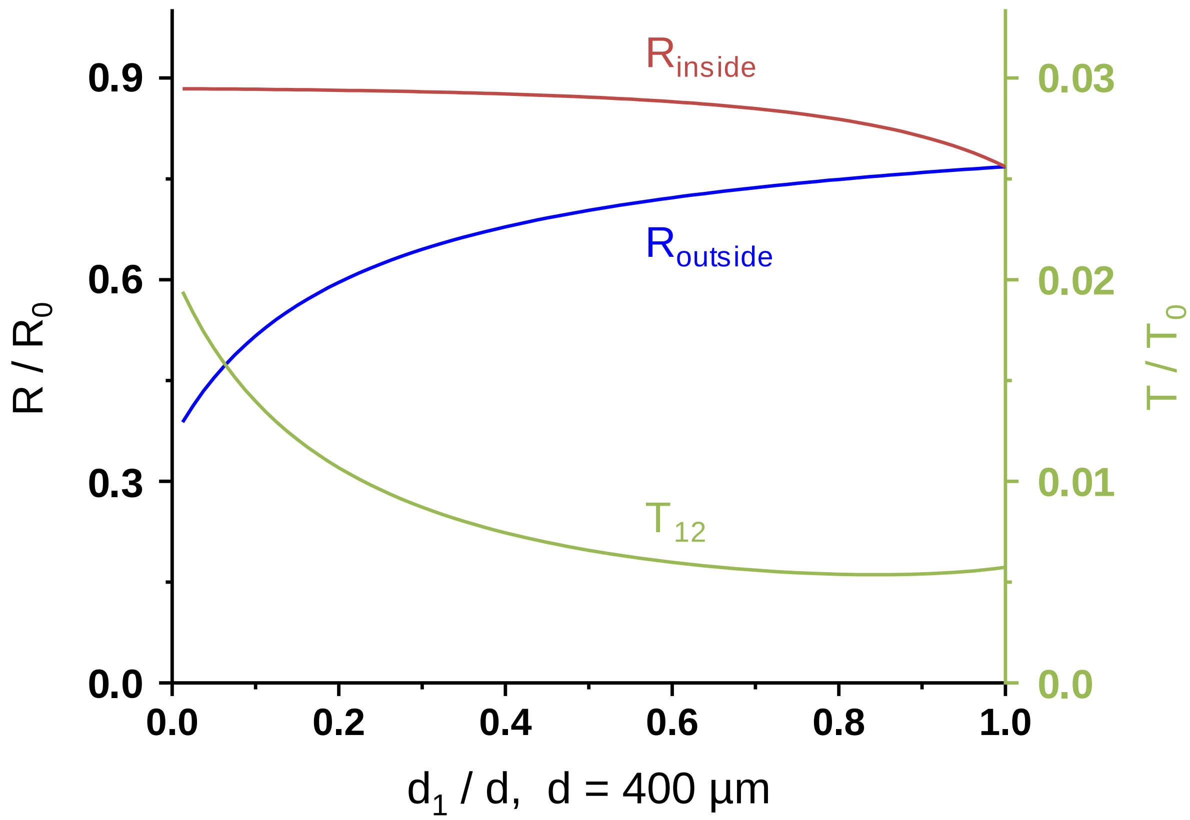
The two layers are added to a bi-layer with a constant total thickness $d = d_1 + d_2$, but variable parts. The total amount of absorber is constant = $K_d = K_1d_1 + K_2d_2$. We start from a uniform absorbing layer with K and $d = d_1$. Afterwards, we reduce d_1 and simultaneously compress the fraction x of the absorber into the same direction. The other parameters remain unchanged. The resulting absorption coefficients are $K_1 = x K d/d_1 + (1 – x) K$, and $K_2 = (1 – x) K$. All relations are inserted in Eqs. (2) of the main article, wherefrom $R_{12} = R_{outside}$, $R_{21} = R_{inside}$, and $T_{12} = T_{21}$ are obtained. Fig.11 shows the results for a data set (d = 400 µm, S = 400 cm^{-1}, K = 14 cm^{-1}, x = 0.8) that is approximately able to reproduce the experiments of the brown shell in Tab. 4. Some features are noteworthy to be mentioned:

The T-curve depends strongly on the absorber compression so that it can help to determine the thickness of the outer layer d_1. This behavior is different from a transparent layer, where the transmittance is independent of the absorber compression according to Beer's law.

The R_{inside} – curve depends almost exclusively on K_2 in the range of experimental interest, $d_1/d < 0.2$.

The formalism uses four variables (S, K, d_1, x) but delivers only three independent experiments. We additionally determine the scattering coefficients in the uniform layer approximation from measurements out of absorption.

The calculated thickness of the strongly absorbing outer layer, $d_1 \approx 15$ µm, is consistent with SEM micrographs and our own ablation experiments. However, the calculated absorption coefficients $K_1 \approx 300$ cm^{-1} and $K_2 = 2.8$ cm^{-1} are far away from reality. The discrepancy is attributed in the main article to repeated Fresnel reflections at the internal shell boundaries that increase the path length of radiation and thus the absorption by about a factor of two.



Supplementary Fig. 1: Diffuse reflectance and transmittance of an absorbing and multiple scattering bi-layer as function of the absorber distribution over the two sublayers. Absorber concentration in sublayer 1: $K_1 = K(xd/d_1 + 1 - x)$, in sublayer 2: $K_2 = K(1 - x)$. Data set for the figure: K = 14, S = 400, x = 0.8, d = 0.04.