Original Paper

Spectral transmittance of Christiansen filters – Experimental observations[1)]

Hans-Jürgen Hoffmann and Ralf Steinhart[2)]

Institut für Nichtmetallische Werkstoffe: Glaswerkstoffe, Technische Universität, Berlin (Germany)

Powder of the optical glass K5 has been immersed into methyl benzoate as refractice index matching fluid to fabricate Christiansen filters. The internal spectral transmittance of these filters has been investigated in the visible spectral region as a function of the filter thickness, the mean diameter of the powder grains, and the difference between the refractive indices of the fluid and the K5 glass.

The minimum internal spectral extinction of the filter curve is approximately proportional to the thickness of the filter. Furthermore, it scales inversely with the average diameter of the glass grains. Hence, one can deduce that the minimum spectral extinction is proportional to the total interface area between grains and immersion liquid. According to the experimental results it can be concluded further that this extinction is mainly due to Rayleigh scattering.

The halfwidth of the spectral transmission passband decreases with increasing thickness of the filters and with decreasing average diameter of the grains. The spectral extinction at wavelengths sufficiently far from its minimum increases sublinearly with the filter thickness and the inverse mean diameter of the grains. In the same spectral region, the extinction increases also sublinearly with the absolute difference between the refractive indices of the material of the grains and of the immersion liquid. Until now, a theory predicting all of these observations correctly seems to be still missing.

Spektrales Transmissionsvermögen von Christiansen-Filtern – Experimentelle Beobachtungen

Aus Pulver des optischen Glases K5 und Methylbenzoat als Immersionsflüssigkeit wurden Christiansen-Filter hergestellt. Das interne spektrale Transmissionsvermögen dieser Filter wurde im sichtbaren Spektralbereich in Abhängigkeit von der Filterdicke, dem mittleren Korndurchmesser und der Brechzahldifferenz zwischen der Immersionsflüssigkeit und dem K5 Glas untersucht.

Das minimale spektrale Extinktionsvermögen der Durchlaßkurve ist näherungsweise proportional zur Filterdicke und umgekehrt proportional zum mittleren Durchmesser der Körner. Hieraus ergibt sich, daß das minimale spektrale Extinktionsvermögen proportional zur Grenzfläche zwischen allen Körnern und der Immersionsflüssigkeit ist. Die experimentellen Ergebnisse können dadurch erklärt werden, daß Rayleigh-Streuung die wichtigste Ursache für diese Restextinktion ist.

Bei konstantem Füllfaktor (d.h. konstantem Verhältnis der Mengen an Glas und Immersionsflüssigkeit) nimmt mit zunehmender Dicke und mit abnehmendem mittlerem Korndurchmesser die Halbwertsbreite der spektralen Transmissionskurve ab. Für Wellenlängenbereiche hinreichend weit weg vom Minimum der Extinktionskurve wächst die spektrale Extinktion sublinear mit der Filterdicke, mit dem Kehrwert des mittleren Korndurchmessers und mit der Brechzahldifferenz zwischen den Glaskörnern und der Immersionsflüssigkeit an. Bis jetzt ist keine Theorie bekannt, die all diese experimentellen Beobachtungen widerspruchsfrei erklärt.

1. Introduction

The spectral transmittance of an inhomogeneous medium which consists of a powder of a transparent solid material immersed in a different material such as a liquid depends on the difference of the refractive indices of both materials. If the refractive indices coincide, the spectral transmittance is maximum. With increasing absolute difference of the refractive indices, the spectral transmittance of a direct beam of electromagnetic radiation decreases. This is the origin of a bell-shaped spectral transmittance curve of the inhomogeneous medium. A plane-parallel layer of such a medium is called Christiansen filter to honour the author who described this effect more than 100 years ago [1and 2].

In the past, a large number of different theories and formulae have been published to describe the spectral transmittance of Christiansen filters. In a recent review paper, Hense [3 and 4] published a thorough survey on the different approaches. Unfortunately, most of the theoretical results do not coincide in the predictions of the spectral transmittance as a function of the filter thickness, the average diameter of the grains or the difference of the refractive indices between the material of the grains and the immersion liquid. Hense, however, did not compare the theoretical predictions with experimental results in detail. Therefore, the aim of the present work was to investigate the spectral transmittance of

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[1)] This experimental work was done when both authors were with Schott Glas, Mainz (Germany).

[2)] Now with Robert Bosch GmbH, Stuttgart (Germany).

Christiansen filters as a function of these parameters experimentally for comparison with the different theoretical approaches and to decide – at least experimentally – which formula was useful and which was not. In section 2, the experimental details of the investigation are described. Then, the experimental results together with the details of the evaluation are presented. Finally, the results are compared with the predictions of a frequently used formula of the internal spectral transmittance of Christiansen filters. Since this formula fails to describe the results correctly, the reasons for this failure are discussed. The authors must point out, however, that until now they are not aware of a theoretical approach from the literature that agrees with all of their experimental observations.

2. Experimental details

For the fabrication of the glass powder a large homogeneous block of the optical glass K5 [5] was used. Part of this block was broken into smaller pieces with maximum diameter of about 1 cm. These pieces were milled in a ball mill into powder. The powder was sieved to form four fractions of different grain ranges: 0.13 to 0.25 mm, 0.25 to 0.5 mm, 0.5 to 1.0 mm, and 1.6 to 2.0 mm with the average diameters $\bar{d}$ = 0.19, 0.38, 0.75, and 1.8 mm.

Particles of iron which could have been introduced into the powder were removed by a magnet. Each fraction of the powder was washed several times in deionized water and acetone (p.a. quality) to remove dust particles. After the washing, the powder was dried at 150 °C for 1 h in a laboratory oven at ambient atmosphere.

The refractive index of the K5 glass was determined from the angle of minimum deviation of a prism cut from the same glass block that was used for the powder. These measurements were done for the standard wavelengths λ = 643.8, 587.6, 546.1, 480.0, and 435.8 nm at 20 °C and three additional temperatures. The data of the refractive index were recalculated to standard atmospheric parameters, i.e. dry air at a pressure of 0.101325 · 10^6 Pa at 20 °C.

These data of the refractive index are fitted by the one-term Sellmeier equation

$$n^2(\lambda, 20\,°\mathrm{C}) - 1 = C_\mathrm{m} \frac{\lambda^2 \cdot \lambda_0^2}{\lambda^2 - \lambda_0^2} \tag{1}$$

with the parameters $C_\mathrm{m} = C_\mathrm{gl} = 1.2544 \cdot 10^{-4}\,\mathrm{nm}^{-2}$ and $\lambda_0 = \lambda_{0,\mathrm{gl}} = 100.96$ nm (the index "gl" stands for K5 glass).

The temperature coefficient $\mathrm{d}n/\mathrm{d}T$ of the refractive index relative to standard air was 1.6 · $10^{-6}\,\mathrm{K}^{-1}$ for 587.6 nm (helium d-line). However, small corrections of the refractive index of K5 glass could be neglected due to variations of the ambient temperature, since the temperature coefficient of the refractive index of the immersion liquid was several orders of magnitude larger than that of the glass.

The immersion liquid was methyl benzoate ($C_8H_8O_2$). The following data on the refractive index of methyl benzoate have been published in the literature: n = 1.518 for the temperature 16.6 °C without specifying the wavelength [6], n = 1.515 for λ = 589.6 nm without specification of the temperature [7], and n = 1.516 to 1.518 at 20 °C for λ = 589.6 nm [8]. Since these data were not sufficiently accurate for this investigation, the authors determined the refractive index of the immersion liquid for the wavelengths 435.8, 480.0, 546.1, 587.6, and 643.8 nm at 20 °C and several additional temperatures between −22.2 and 43.3 °C. The data at 20 °C were recalculated relative to standard air and fitted by a one-term Sellmeier equation (1) with the fitting parameters $C_\mathrm{fl} = 6.043 \cdot 10^{-5}\,\mathrm{nm}^{-2}$ and $\lambda_{0,\mathrm{fl}} = 142.37$ nm. (The index "fl" stands for the "fluid" methyl benzoate). The decrements and increments of the refractive index for different temperatures are fitted by

$$\Delta n_\mathrm{fl}(\lambda, \Delta T) = \frac{n_\mathrm{fl}^2(\lambda, 20\,°\mathrm{C}) - 1}{2 \cdot n_\mathrm{fl}(\lambda, 20\,°\mathrm{C})} \left(A \cdot \Delta T + \frac{B \cdot \Delta T}{\lambda^2 - \lambda_{0,\mathrm{fl}}^2} \right) \tag{2}$$

wherein ΔT is the temperature difference with respect to 20 °C, $A = -1.1143 \cdot 10^{-3}\,\mathrm{K}^{-1}$, and $B = -3.4668 \cdot 10^{-3}\,\mathrm{K}^{-1}\,\mathrm{nm}^2$. Thus, the refractive index of the liquid is given for the wavelength λ and the temperature T = 20 °C + ΔT by

$$n_\mathrm{fl}(\lambda, T) = n_\mathrm{fl}(\lambda, 20\,°\mathrm{C}) + \Delta n(\lambda, \Delta T). \tag{3}$$

To fabricate the Christiansen filters cuvettes with inner cross section of (20 × 20) mm² were used. These cuvettes were made of 1 mm thick plates of K5 glass. Glass powder was immersed in small quantities into the cuvettes filled partly with methyl benzoate. To avoid small bubbles of gas, the cuvettes were tapped during this procedure. The fill factor, i.e. the ratio of the volume of the glass powder to the volume of both the powder and the liquid, was f = 0.5. This was determined by weighting before and after filling the cuvettes with immersion liquid (mass density: 1.086 g/cm³) and glass powder (mass density: 2.59 g/cm³). The thickness of the Christiansen filters was modified by inserting plane-parallel plates of K5 glass into the cuvettes. These plates were 10, 15, and 17 mm thick, resulting in 3, 5, 10, and 20 mm as the effective thickness of the filters.

The spectral transmittance of the Christiansen filters was determined using a two-beam spectrometer Lambda 9 from Perkin Elmer. The light source was a tungsten halogen lamp. To obtain monochromatic radiation, the spectrometer has a built-in filter wheel with suitable passband filters and a double grating monochromator. The light was modulated in the spectrometer by a chopper. During one half-period the beam passed through the sample and during the other half-period it passed

through the reference, which consisted of a 22 mm thick K5 plate. Thus, the transmittance of the Christiansen filters was corrected for the reflection losses on the surfaces automatically yielding the internal spectral transmittance and the internal spectral extinction. (Throughout the present publication internal spectral quantities were used.) In addition, differences in the optical path length between the reference and the measuring beam were avoided. The full aperture angle was reduced by a diaphragma to 12° in the vertical direction and to 8° in the horizontal direction. The detector consisted of an Ulbricht sphere with a standard photomultiplier. The detector aperture remained constant during the experiments. It was fully illuminated with K5 plates of 22 mm thickness in both the references and the sample beam.

Since the spectral transmittance maximum of the Christiansen filters shifts rather sensitively with the temperature (due to the large variation of the refractive index of the immersion liquid with the temperature given by equation (2)), the authors had to control the temperature of the filters very accurately. Therefore, the filters were kept in the measurement compartment of the spectrometer and the instrument was not switched off during the period of the experimental investigations. The temperature of the filters was 27.1 °C.

The spectrometer was controlled by a PC using the software PE CSS1 from Perkin Elmer. The experimental results were registered as both spectral transmittance, $I_f(\lambda)/I_0(\lambda)$, and spectral optical density or spectral decadic extinction, $\lg(I_0(\lambda)/I_f(\lambda))$, wherein $I_0(\lambda)$ and $I_f(\lambda)$ are the intensities of the light beams through the reference and the Christiansen filters. (Throughout the paper "spectral extinction" is used for the spectral decadic extinction coefficient for short.) The time constant to reach 98 % of the final values was 0.5 s. The scanning speed for the spectrum was chosen 240 nm/min; the total spectral resolving width was about 4 nm.

3. Experimental results and their evaluation

Figures 1 and 2 show the internal spectral transmittance $I_f(\lambda)/I_0(\lambda)$ and the internal spectral extinction $\lg(I_0(\lambda)/I_f(\lambda))$ of Christiansen filters with a path length of z = 20 mm for different average grain diameters $\bar{d}$. The internal spectral transmittance of the Christiansen filters (K5 glass in methyl benzoate) is maximum at the wavelength λ = 462 nm for the temperature T = 27.1 °C. The full width of the passband at half maximum decreases with decreasing grain size of the glass powder, as can be seen from figure 1. The spectral extinction minimum seen in figure 2 increases with decreasing mean diameter of the glass grains $\bar{d}$.

The internal spectral transmittance and the internal spectral extinction for the average grain size $\bar{d}$ = 0.75 mm is shown in figures 3 and 4 for different path lengths z. Both the maximum of the internal transmittance and the width of the passband decrease with increasing path length z (figure 3), whereas the internal

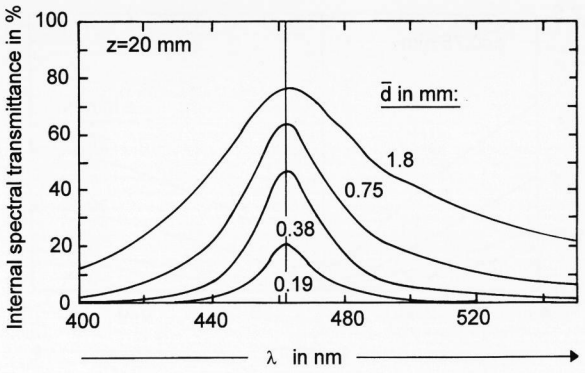

Figure 1. Internal spectral transmittance as a function of wavelength λ for different average grain size $\bar{d}$ and constant thickness z = 20 mm of the Christiansen filters.

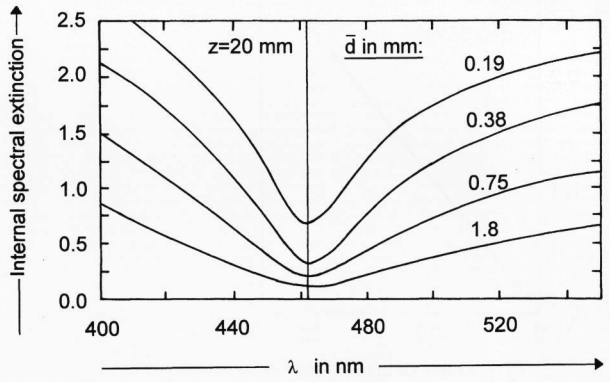

Figure 2. Internal spectral extinction of Christiansen filters as a function of wavelength λ for different average grain size $\bar{d}$ and constant thickness z = 20 mm.

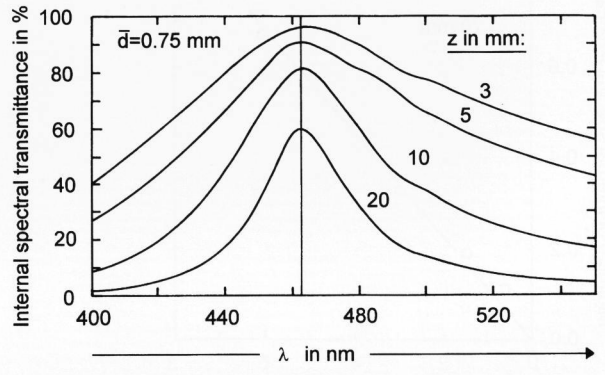

Figure 3. Internal spectral transmittance as a function of wavelength λ for different thickness z of the Christiansen filters and constant average diameter of the grains $\bar{d}$ = 0.75 mm.

spectral extinction increases with z (figure 4). This behaviour corresponds to that shown in figures 1 and 2 for a given path length and different grain size. This correspondence suggests that the internal spectral transmittance is rather a function of the number of interfaces between the two phases (one phase is the liquid and the

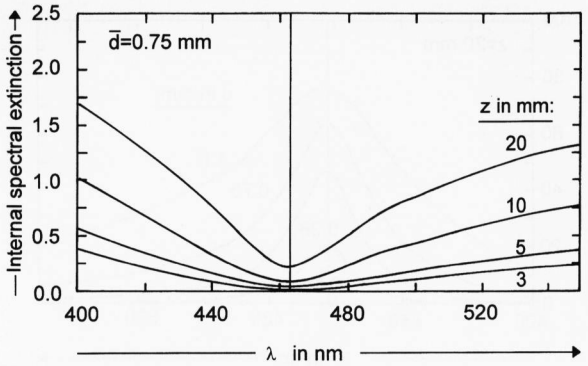

Figure 4. Internal spectral extinction as a function of wavelength λ for different thickness z of the Christiansen filters and constant average diameter of the grains $\bar{d}$ = 0.75 mm.

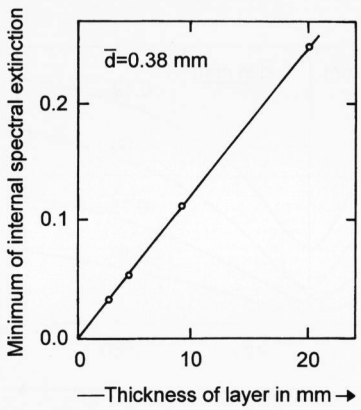

Figure 5. Minimum of the internal spectral extinction as a function of the thickness z (average grain size $\bar{d}$ = 0.38 mm).

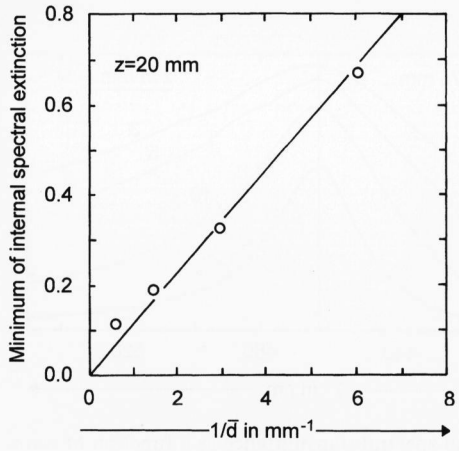

Figure 6. Minimum of the internal spectral extinction as a function of the reciprocal average diameter $\bar{d}$ of the grains for thickness z = 20 mm of the Christiansen filters.

other is the glass) of the Christiansen filters. Geometrical similarity can obviously be taken for granted in this case if

(a) the cross section of the light beam is much larger than the average cross section of the grains,

b) the shape of the grains is statistically similar,

c) internal loss mechanisms of the materials of the two phases, which depend on the path length in each of both phases separately, can be neglected, and

d) the wavelength is very small as compared to the geometric dimensions of the grains.

With these conditions for two given materials, the Christiansen filters should behave similarly from a point of view of geometrical optics, if $z/\bar{d}$ is invariant. If the average diameter of the grains is in the order of the wavelength of the electromagnetic radiation, the conditions for this statement are certainly no longer fulfilled, since diffraction has to be taken into account.

The scaling law of the internal spectral extinction with z or $\bar{d}$ alone is not obvious. However, the authors observed that its minimum value (see figure 4) is proportional to the thickness z of the filters at a given average grain size. This can be seen in detail from figure 5, which shows the minimum of the internal spectral extinction being proportional to z for a given average diameter $\bar{d}$. Since the internal spectral transmittance and the internal spectral extinction depend on the ratio $z/\bar{d}$, as has been shown above already, the minimum value of the internal spectral extinction is expected to be proportional to the inverse of $\bar{d}$ for a given thickness z of the filters. The experimental results shown in figure 6 agree with this expectation.

The internal spectral transmission maximum should be 100 % or the minimum of the internal spectral extinction should be zero, because in this case the refractive index of the glass grains should match that of the immersion liquid (see [9], e.g.). The experimental observations (see figures 1 to 4), however, do not agree with this expectation. Therefore, one must assume that either at least one additional loss mechanism is active even without a difference of the refractive indices of both materials or the refractive indices do not match. Generally, any inhomogeneity of a material can cause differences in the refractive indices and in the polarizability of the ions or molecules. Inhomogeneities within the glass is one possible explanation. Another possibility is given by small changes of the polarizability of the ions and molecules at the interface glass/immersion liquid either in the surface areas or at the edges or at the corners. In each of these cases, the dependence of the loss on the average diameter $\bar{d}$ is different. Inhomogeneities of the refractive index in the bulk of the glass sample scale with the volume of the grains, i.e. as $\bar{d}^3$. The interface area of a grain scales as $\bar{d}^2$, the edges as $\bar{d}^1$ and the average number of corners is fixed and scales as $\bar{d}^0$. In order to obtain a quantity proportional to the internal spectral extinction at its minimum, this has to be multiplied by the total number of grains per volume, which scales as $1/\bar{d}^3$ for a constant fill factor. Thus, in total the minimum internal spectral extinction would be proportional to $\bar{d}^0$, i.e. independent of the grain size, if the loss is proportional to inhomogeneities distributed in the bulk glass, or proportional to $1/\bar{d}$, if its origin is due to the surface areas, or pro-

portional to $1/\bar{d}^2$, if it is due to the edges, and proportional to $1/\bar{d}^3$, if the loss is caused by the corners. The experimental results in figure 6 show clearly that the loss at the minimum of the internal spectral extinction is proportional to $1/\bar{d}$, i.e. it is due to some mechanism effective in the surface areas.

The authors tentatively assumed that this loss can be decribed by a term constant/λ^4 due to Rayleigh scattering of microinhomogeneities smaller than the wavelength created by the milling and by the leaching of the surface in the washing process of the powder. Rayleigh scattering scales as $1/\lambda_0^4$. If this mechanism contributes to the extinction, the minimum internal spectral extinction would be proportional to $1/\lambda_0^4$ with λ_0 being the wavelength, for which this minimum is observed. For this reason the refractive index of the immersion liquid was varied by choosing different fluids and the internal spectral extinction minimum shifted. Figure 7 shows that the value of the internal spectral extinction minimum is a linear function of $1/\lambda_0^4$ confirming that Rayleigh scattering contributes to the extinction.

In [10] data are given on the maximum of the external spectral transmittance T of Christiansen filters as a function of wavelength λ_0 if the refractive index of the immersion liquid was changed. From these data were calculated the corresponding data of the minimum external spectral extinction, $\lg(1/T)$, which have been plotted in figure 8 as a function of $1/\lambda_0^4$ (with λ_0 being the wavelength for which the minimum has been observed). One can clearly see that the minimum external extinction is a linear function of $1/\lambda_0^4$ indicated by the solid line in figure 8. This is in agreement with the observation that Rayleigh scattering contributes to the transmission losses of Christiansen filters. (The abscissa on the vertical axis for $1/\lambda_0^4 \rightarrow 0$ in figure 8 corresponds to reflectance losses of the surfaces of the filters and possible other losses, which do not vary appreciably with the wavelength in the range of the experimental data.)

Further support to this result arises form the asymmetry of the internal spectral extinction curves to be dealt with in more details in the following: In order to compare the experimental results with the predictions of several formulae published in the literature, the internal spectral extinction was represented as a function of the absolute difference of the refractive indices $|\Delta n|$ between the glass and the immersion liquid. Figure 9 shows some examples. For wavelengths longer (lw) than the wavelength λ_0 of the internal spectral extinction minimum at 462 nm, the refractive index of the glass is larger than that of the immersion fluid, whereas for shorter wavelengths (sw) the opposite is true. The curves in figure 9 show that the internal spectral extinction is nearly independent of $|\Delta n|$ near the minimum; in the neighbouring range it is roughly proportional to $|\Delta n|$ and for even larger values of $|\Delta n|$ it increases sublinearly with $|\Delta n|$, i.e. it increases less than proportional to $|\Delta n|$. For the same values of $|\Delta n|$, however, the internal spectral extinction is smaller on the long wavelength side as compared to the short wavelength side. This asymmetry can be attributed to different contributons of the Rayleigh scattering, which increases with decreasing wavelength. In this case, the difference of the internal spectral extinction data on both sides should be proportional to $\frac{1}{\lambda_{sw}^4} - \frac{1}{\lambda_{lw}^4}$, wherein λ_{sw} and λ_{lw} are corresponding wavelengths of the same $|\Delta n|$ on the short and long wavelength side of the internal spectral extinction curve. In figure 10, examples are shown of the differences of the experimental internal spectral extinction curves as a function of the corresponding $\frac{1}{\lambda_{sw}^4} - \frac{1}{\lambda_{lw}^4}$. (The data in the vicinity of the extinction minimum have been omitted, since the error bars due to small differences of the spectral extinction would be too large.) In fact, this difference is proportional to $\frac{1}{\lambda_{sw}^4} - \frac{1}{\lambda_{lw}^4}$, thus showing

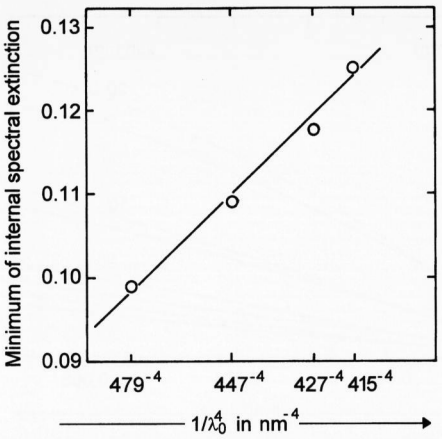

Figure 7. Minimum of the internal spectral extinction of Christiansen filters (thickness z = 20 mm, average grain size $\bar{d}$ = 1.8 mm) as a function of $1/\lambda_0^4$. The minimum of the spectral extinction has been shifted by changing the refractive index of the immersion liquid.

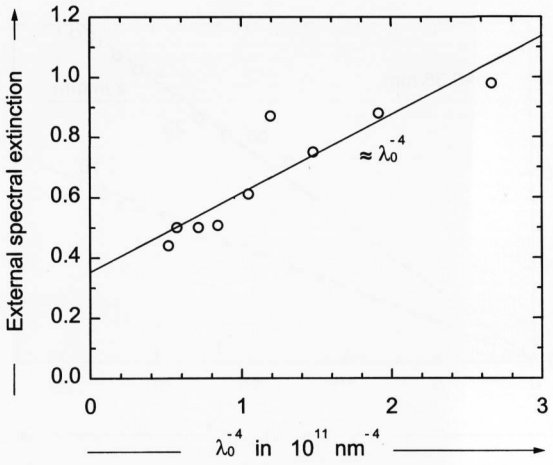

Figure 8. Minimum external spectral extinction as a function of $1/\lambda_0^4$. The data are taken from [10].

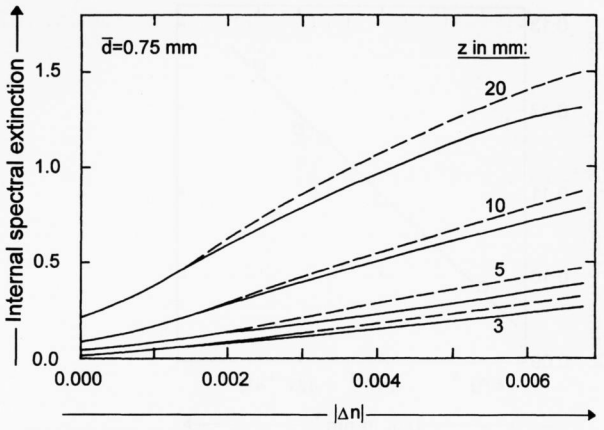

Figure 9. Internal spectral extinction as a function of the absolute difference $|\Delta n|$ of the refractive indices between glass and immersion liquid. (The solid and dashed curves refer to the long and short wavelength side – "*lw*" and "*sw*" –, respectively, of the spectral transmission curves.) Average grain size: $\bar{d}$ = 0.75 mm.

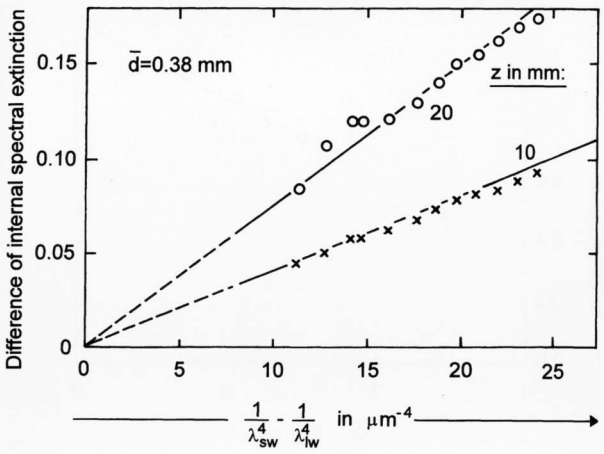

Figure 10. Difference of the internal spectral extinction on the short and long wavelength side for the same $|\Delta n|$ as a function of the appertaining $1/\lambda_{sw}^4 - 1/\lambda_{lw}^4$ (z = 20 mm, $\bar{d}$ = 0.38 mm).

that Rayleigh scattering can cause the asymmetry of the internal spectral extinction curves of the Christiansen filters.

4. Discussion

The optical glass K5 (crown glass family) is an alkali silicate glass with small amounts of B_2O_3. Since alkali silicate glasses can show phase separation, one might assume that the observed Rayleigh scattering is due to the onset of such a phase separation. Phase separation in the volume as the cause for the scattering, however, can definitively be ruled out for the following reason: the losses due to Rayleigh scattering scale in the present case with the inverse of the average diameter $\bar{d}$ of the grains. As has been shown in the preceding section, this corresponds to a scaling with the interface area between the grains and the immersion liquid. In the case of phase separation in the bulk, the scattering losses would be independent of $\bar{d}$. At present the true cause for the surface scattering is not known. One possible reason may be found in the treatment of the powder: the washing with water or other polar solvents may attack the surface of the glass, since optical glasses are generally not as stable against chemical attack as technical glasses. It may be worthwhile continuing these investigations to study the influence of different solutions and various treatments on the glass powder.

In the present investigation the main emphasis, however, was laid on the evaluation of the data of the internal spectral transmittance of the Christiansen filters under several aspects for comparison with theoretical predictions of different formulae known from the literature. To serve as selective criteria it is worthwhile summing up the main presumptions and the main results of the present experimental investigation. The diameter of the glass grains is presumed to be much larger than the wavelength of the electromagnetic wave and much smaller than the diameter of the light beam. Furthermore, loss mechanisms in the bulk glass and in the liquid should be negligible. The dominant loss should be due to the interface between the glass grains and the immersion liquid. The glass and the immersion liquid are assumed to be optically homogeneous without any deliberate inhomogeneity of the refractive indices. The interval of the diameter of the glass grains is reasonably small and the temperature of the Christiansen filter remains constant during the experiments. The aperture angle of the measuring equipment must be defined and should be constant.

With these presumptions the following experimental results of the investigation can be generalized:

a) The internal spectral transmittance, $I_f(\lambda)/I_0(\lambda)$, and the internal spectral extinction, $\lg(I_0(\lambda)/I_f(\lambda))$, are functions of $z/\bar{d}$, i.e. of the number of interfaces in direction of the propagation of the light beam.

b) The internal spectral extinction near its minimum is nearly independent of the difference of the refractive index between glass and immersion. In addition, it is larger than zero and it is proportional to the thickness of the filter, z, and to the inverse of the average diameter, $1/\bar{d}$, of the grains.

c) If other loss mechanisms can be excluded, the internal spectral extinction at its minimum is caused mainly by Rayleigh scattering. Consequently, it is proportional to the total Rayleigh scattering intensity, which scales with the wavelength as $1/\lambda^4$.

d) With increasing absolute difference of the refractive indices $|\Delta n|$ between the glass and the immersion liquid the internal spectral extinction increases rather sublinearly with $|\Delta n|$, the thickness z of the filters, and the reciprocal average diameter of the grains $1/\bar{d}$.

e) The full spectral width of the internal spectral transmission curves decreases with increasing $z/\bar{d}$ (not necessarily proportional to $z/\bar{d}$, however).

Comparing the theoretical predictions of the formulae compiled in [3 and 4], it seems that at present there is no theory compatible with the experimental results shown above. As an example the authors discuss in some detail the discrepancies with respect to the formulae

$$\tau_0 = \exp\{-k^2\pi^2\lambda^{-2}\bar{d}\,z\,\Delta n^2\} \tag{4}$$

for the internal spectral transmittance [9] and the resulting internal spectral extinction

$$E = k^2\pi^2\lambda^{-2}\bar{d}\,z\,\Delta n^2 \lg e \tag{5}$$

(k^2 = constant; λ = wavelength of the electromagnetic radiation; $\bar{d}$ = average diameter of the glass grains; z = thickness of the filter; Δn = difference of the refractive indices between the glass and the immersion liquid). Such a comparative test seems to be worthwhile since equation (4) is the basis of a homogeneity test for glasses [11]. According to equations (4) and (5), the maximum internal transmittance should be 100% for $\Delta n = 0$, and the full spectral width at half maximum should increase with decreasing average diameter. In addition, the internal spectral extinction should increase with the square of Δn and increase proportional to the filter thickness z.

None of these predictions agrees with the experimental observations. The maximum internal spectral transmittance never reaches 100%. The full spectral width of the transmittance curve does not increase, but decreases with decreasing $\bar{d}$; the internal spectral extinction increases with increasing inverse of $\bar{d}$ and increases much less than proportional to Δn^2, rather as Δn at most. Furthermore, it increases sublinearly with z. Similar discrepancies exist between the other formulae cited in [3 and 4] and the experimental results, too. Thus, a theory describing the spectral transmittance and the spectral extinction of Christiansen filters correctly (at least within reasonable approximations) seems to be still missing, despite a very large number of experimental and theoretical publications on that topic.

Finally, the authors want to add some remarks on the failure of equations (4) and (5) to describe the experimental results, since discrepancies to formula (5) have been observed also by other researchers in this field. Afghan and Cable [12] reported that their experimental results could be fitted rather by

$$E = k'\,\Delta n^{1.85} z^1 \lambda^0/\bar{d}\,. \tag{6}$$

This is in contrast to equation (5) with respect to the dependence on $\bar{d}$ and λ. These authors also reported on an asymmetry of the transmission passband similar to that presented here. Unfortunately, their results refer to the external spectral extinction. Thus, a quantitative comparison of their data points with the present results is not allowed.

The applicability of Raman's waveoptical theory to Christiansen filters has been questioned also by Varshneya [13]. He presumes that Raman's theory is applicable to glass particles less than 30 µm in size, whereas for larger particles geometrical optics has to be applied. Raman's theory, however, seems to be oversimplified, since he takes into acount in his formula (equation (4) above) the interference effects due to phase shifts between the glass grains and the surrounding immersion liquid for a coherent plane wave and for zero aperture angle, only. Using a spectrometer in practice, the electromagnetic wave is incoherent and the aperture angle is larger than zero. For an incoherent wave with larger aperture angle the contribution considered by Raman cannot account for the spectral transmission curve. On the other hand, he neglected the following in his theory:

- reflection losses at the boundaries between the grains and the immersion liquid;
- scattering losses, such as Rayleigh scattering;
- diffraction losses on the (statistical) phase structure grains/immersion liquid; and
- deflection (refraction) losses due to the multiple prismatic shape of the grains.

In this context losses mean the deviation of electromagnetic power out of the aperture angle of the incident beam due to the Christiansen filter. A realistic theory of such filters has to take account of all of these effects.

In this respect one has to consider the Monte Carlo approach performed by Varshneya, Loo, and Soules [14] for the spectral transmittance of Christiansen filters using geometrical optics. These authors took account of reflection losses and refraction by the grains. They neglected scattering, as has been detected in the present work, and diffraction on the statistical phase structure of the filters. Since reflection and refraction seem to be the dominant effects (also to the present authors' own observations), Varshneya et al. [14] obtained rather fair agreement between their experimental results and their numerical statistical calculations. They applied the statistical calculations to powder of inhomogeneous glasses sucessfully, too. However, they did not make any statement about the dependence of the transmittance curves on the thickness of the filters, the average diameter of the grains, and the difference between the refractive indices. In addition, an explicit formula describing the transmittance of Christiansen filters obviously cannot be obtained from Monte Carlo calculations.

5. References

[1] Christiansen, C.: Untersuchungen über die optischen Eigenschaften von fein vertheilten Körpern. T. 1. Ann. Phys. Chem., ser. 3, **23** (1884) No. 10, p. 298–306.

[2] Christiansen, C.: Untersuchungen über die optischen Eigenschaften von fein vertheilten Körpern. T. 2. Ann. Phys. Chem., ser. 3, **24** (1885) No. 3, p. 439–446.

[3] Hense, C. R.: The Christiansen filter – A centennial retrospective review. Part 1. Glastech. Ber. **60** (1987) no. 3, p. 89–111.

[4] Hense, C. R.: The Christiansen filter – A centennial retrospective review. Part 2. Glastech. Ber. **60** (1987) no. 4, p. 140–160.

[5] Optical glass. Catalogue no. 10.000, 1992. Schott Glas, Mainz (Germany).
[6] Weigert, F.; Staude, H.: Über monochromatische Farbfilter. Z. Phys. Chem. **A 130** (1927) p. 607–615.
[7] Lusty, M. E.; Dunn, M. H.: Refractive indices and thermo-optical properties of dye laser solvents. Appl. Phys. **B 44** (1987) p. 193–198.
[8] Neumüller, O. A. (ed.): Römpps Chemie-Lexikon, 7th ed. Stuttgart: Franckh, 1972. p. 336.
[9] Raman, C. V.: The theory of the Christiansen experiment. Proc. Indian. Acad. Sci. **A 29** (1949) no. 5, p. 381–390.
[10] Elvegård, E.; Staude, H.; Weigert, F.: Über monochromatische Lichtfilter. T. 2. Z. Phys. Chem. **B 2** (1929) no. 3, p. 149–160.
[11] Shelyubskii, V. I.: A new method for determining and controlling the homogeneity of glass. (Orig. russ.) Steklo Keram. **17** (1960) no. 8, p. 17–22.
[12] Afghan M.; Cable, M.: Estimation of the homogeneity of glass by the Christiansen filter method. J. Non-Cryst. Solids **38/39** (1980) p. 3–8.
[13] Varshneya, A. K.: Glass inhomogeneity measurement by Shelyubskii technique – A criticism of Raman's Christiansen filter theory. In: XIV International Congress on Glass, New Delhi 1986. Collected papers. Vol. II. Calcutta: Indian Ceram. Soc. 1986. p. 271–279.
[14] Varshneya, A. K.; Loo, M. C.; Soules, T. F.: Glass inhomogeneity measurement using the Shelyubskii method. J. Am. Ceram. Soc. **68** (1985) no. 7, p. 380–385.

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Addresses of the authors:

H.-J. Hoffmann
Institut für Nichtmetallische Werkstoffe:
Glaswerkstoffe
Technische Universität
Englische Straße 20
D-10587 Berlin

R. Steinhart
Robert Bosch GmbH
Postfach 106 050
D-70049 Stuttgart