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Analysis of the dispersion of optical plastic materials ☆

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1. Introduction

At present plastic materials find wide application in consumer and high quality optics. Optical plastics (OPs) are used mainly in the visible (VIS) and near-infrared (NIR) spectral regions from 400 nm to 1100 nm. Success in application of OPs depends on knowledge of their optical refraction, transmission, birefringence, haze and homogeneity [1]. The optical properties of polymers are in details considered in [2]. Chromatic dispersion is an important characteristic in the design of optical systems and devices. However, measurements of refractive indices are usually realized at several selected wavelengths. Determination of more extensive refractometric data is possible using dispersion formulae [3,4].

The measuring methods for determination of OPs' refractometric characteristic are quite different. The refractive indices of transparent polymers can be obtained using the Federal Test Method Standard [5] in which the Abbe refractometer is applied. It operates with a white light source and Amici prisms as colour compensators. The refractive index value for the sodium *D*-line can be read directly from the instrument. However, the measuring accuracy is not acceptable for modern optical design projects. Furthermore, determination of refractive indices values can not be realized at different wavelengths. Utilization of Zeiss Pulfrich refractometer (PR2) is possible too [6,7]. We have measured the refractive indices in the VIS light using the PR2 instrument with its V-type prism [8] and additional goniometric set-up was applied for the entire VIS and NIR regions. The obtained refractive values were compared with the data from Glass catalogues of OSLO [9], ZEMAX [10] and Code V [11]. Laser refractometric measurements of a number of OP specimens have been also accomplished using a He–Ne laser source with 632.8 nm emission wavelength [12].

Most widely used OPs are thermoplastics as polymethyl methacrylate (PMMA), polystyrene (PS), polycarbonate (PC), methyl methacrylate styrene copolymer (NAS), styrene acrylonitrile (SAN) and methylpentene (TPX) [1,13]. The only thermosetting plastic for optical applications is allyl diglycol carbonate (CR-39) [1]. The available catalogue data for refractive indices and dispersion characteristics of OPs is yet scanty. Useful data of commonly used transparent polymers is presented in Refs. [1,13]. The refractive indices of the principal OPs are included in many patents [14–17] and available online web-pages [18,19]. Companies producing trade-marks of optical polymers provide information on their refractometric and dispersive characteristics [20]. In comparison with glass, OPs have a restricted range of refractive indices and dispersion. The magnitude of the refractive index n_D at the sodium *D*-line (589.3 nm) usually varies from 1.47 to 1.59 [1,21]. The Abbe number of OPs is in the range from about 100 to a little less than 20 [1,21]. However, there are some differences in the reported refractometric data. For example, the refractive indices values of such a popular material as PMMA are different in references [15,16].

Recently it has been noticed considerable interest in the development of OP materials. Plastics replace glasses in products as objectives, lens arrays, aspheric and ophthalmic lenses, displays and lighting fixtures, windows, internally illuminated outdoor signs and skylights [21,22]. The improvement of manufacturing processes makes possible the utilization of OPs in medicine, military optics, sensors

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and communications [21–24]. Chemical companies produce various trade-marks of OPs as NAS-21 Novacor®, CTE-Richardson®, Zeonex®, Optorez®, Bayer®, etc. but some of them have close dispersion data. Our previous refractometric measurements [7] show for example that Optorez 1330® and S – low Styrene® have similar dispersive properties, Zeonex E48R® and COC® are equal, and Bayer® is a PC-type plastic material. Using some new optical materials the designers can improve the performance and balance the production expenses [13]. It seems that OPs can be both a low-cost alternative to glass and an option that provides more degrees of freedom for product and optical design.

In this paper we consider the refractometric and dispersion properties of OP materials in the region of normal dispersion. We have examined various types of OPs including the principal, selected trade marks and some control samples of polymers.

2. Theoretical analysis of dispersion

The interaction between the electromagnetic wave and the medium (the refractive index n, respectively) depends on its density and individual properties of the molecules on one hand, and the radiation wavelength on the other hand. A characteristic of the materials is the ratio, named a specific refraction $r = f(n)/\rho$, where ρ is the density of the substance and $f(n)$ is a function of the refractive index. The product of the specific refraction and molar mass is the molar refraction R often used in practice. Considering the effective electric field acting on a molecule in a polarizable medium, Lorentz and Lorenz have formulated theoretically [2] the molar refraction R_{LL} as

$$R_{\mathrm{LL}} = \frac{n^2 - 1}{n^2 + 2}\frac{M}{\rho} = \frac{1}{3}N_{\mathrm{A}}\alpha, \qquad (2.1)$$

where M is the molar mass, α is the molecular polarisability and N_{A} is the Avogadro's number. The molar refraction is a material characteristic of the refraction properties which is independent on the density, temperature and physical state. This equation is valid for isotropic materials in cases of ulrta-violet and visible illumination, where the electron polarisability is essential. For organic liquids Gladstone and Dale have obtained that at standard wavelengths the ratio $(n - 1)/\rho$ is a material characteristic constant and the molar refraction is:

$$R_{\mathrm{GD}} = \frac{M}{\rho}(n - 1). \qquad (2.2)$$

Another correlation between the chemical structure of electrically insulating materials and the refractive index has been proposed by Vogel [2]:

$$R_{\mathrm{V}} = nM. \qquad (2.3)$$

It is known that the refraction is an additive value. Therefore, the refractive index can be estimated using Eqs. (2.1), (2.2) and (2.3), if the structure of the compounds of 43 functional groups, using an extensive regression analysis [2]. With his group contributions the refractive index can be predicted with an average standard deviation of about 0.4%.

Variation of the refractive index with respect to the wavelength depends also on the structure of the substance. In the spectral region of transmission the refractive index of the materials reduces towards longer wavelengths. Close to the absorption bands the refractive index enhances with the increase of the wavelength. The major OPs absorb in the blue portion of the visible spectrum and have some energy absorption at wavelengths of 900 nm, 1150 nm and 1350 nm in the NIR region [21]. At longer wavelengths high transmission is possible only in very thin sections of the material (0.022 mm) [1]. OPs become totally opaque at about 2100 nm [21]. The transmission characteristics depend strongly on quenching the material from temperature of about 213 °C to 147 °C in 60 s or less to prevent crystallization [1]. At wavelengths within the absorption bands intramolecular oscillations appear and the bond lengths and valence angles of molecules are altered. The more complex chemical structures of polymers increase the number of absorption bands and therefore influence the dependence of the refractive index on the wavelength.

There are several formulae in literature approximating the dispersion of optical materials. Most popular among them are the Hezberger's experimental formula [3], Cauchy's and Sellmeier's equations [4]. Only the last one, however, has physical ground. Dispersion can be explained by applying the classical electromagnetic theory to the molecular structure of the medium. Sellmeier has considered the substance as a system of elastically bounded particles with natural angular frequency ω_{0i} ($i = 1 \ldots k$ – consecutive number of a single oscillator). The amplitude of the oscillations of bound charges forced by the electromagnetic wave increases at resonance frequency. According to Sellmeier's theory the well-known dependence of the refractive index on the wavelength is obtained:

$$n^2(\lambda) = 1 + \sum_{i=0}^{k} \frac{A_i\lambda^2}{\lambda^2 - \lambda_{0i}^2}, \qquad (2.4)$$

where A_i are constants proportional to the number of oscillators with natural wavelengths λ_{0i} per unit volume.

An important characteristic of the dispersion properties of optical materials is their Abbe numbers, which usually decrease as the refractive indices increase. The USA standard Abbe number v_d is defined by the following ratio:

$$v_d = \frac{n_d - 1}{n_F - n_C}. \qquad (2.5)$$

The difference $(n_F - n_C)$, named a principal dispersion, involves refractive indices n_F and n_C at the blue hydrogen F-line (486.13 nm) and red hydrogen C-line (656.27 nm). The Abbe number v_{804} which is a measure of partial disper-

$$\nu_{804} = \frac{n_{804} - 1}{n_{703} - n_{1052}}, \quad (2.6)$$

where n_{703}, n_{804} and n_{1052} are the refractive indices at wavelengths 703 nm, 804 nm and 1052 nm, respectively. In Europe and Japan the Abbe number is defined according to the green mercury *e*-line (546.07 nm) as:

$$\nu_e = \frac{n_e - 1}{n_{F'} - n_{C'}}, \quad (2.7)$$

where $n_{F'}$ and $n_{C'}$ – refractive indices at the cadmium blue F'-line and red C'-line. Materials with low refractive indices usually have low dispersion behaviour and therefore a high Abbe number.

3. Measurement of the indices of refraction

In this study we apply the measuring method described in details in our paper [7]. The OPs' indices of refraction were measured with the aid of the Carl Zeiss Jena Pulfrich-Refractometer PR2 [8] in the visible spectral region at six standard spectral lines: green *e*-line 546.07 nm, blue *g*-line 435.83 nm, yellow *d*-line 587.56 nm, red *r*-line 706.52 nm, blue *F*-line 486.13 nm and red *C*-line 656.27 nm. We have chosen the *V*-type SF3 glass prism (VoF3 prism) which usually used for measuring liquids since the standard total internal reflection prism requires thick cubic of OP samples with satisfactory polished surfaces to observe a contrast image of the dividing border between the light and dark field in the eyepiece of the instrument. Furthermore, the standard prism does not avoid evaporating of the water contacting solutions and the precision of monitoring decreases at the end of the measuring series.

The examined OP specimens were prepared as injection moulded plates with thickness varying from 2.54 mm to 5.1 mm, except for the control samples which were cubic. Two mutually perpendicular surfaces of the samples were well polished to obtain a good refractometric data. Measuring temperature of 20 °C is maintained by a thermostat and temperature regulation is possible with stability of 0.2 °C. A saturated aqueous solution of zinc chloride ($n_e = 1.51$) and silicon oil ($n_D = 1.56$) for low refractive OP samples as PMMA, and a saturated water solution of potassium–mercuric–iodide (KHgI) with $n_e = 1.73$ for higher refractive PS, PC, etc. have been chosen to ensure the optical contact during the measurements. Initial estimation of the indices of refraction of the OP samples and the choice of immersion emulsions have been accomplished by means of an ellipsometric laser system LEF-3 M-1 made by Carl Zeiss Jena which measuring accuracy of $\Delta n = \pm 0.002$ is completely insufficient to obtain precise OPs' refractometric data.

Refractive indices of OPs in VIS and NIR region are measured with the experimental set-up illustrated in Fig. 1. A G5-LOMO goniometer with an accuracy of one arc second was used with the VoF3 prism-measuring block positioned on the G5 test table. The lighting module operates with a 250 W halogen lamp applied over the entire VIS and NIR regions with interference filters (IF) made by Carl Zeiss (Jena). A new photo detector device was assembled with the aid of a plane silicon diode, operating amplifier and indicating module. The collimator forms a white light beam that falls on the fixed filter. The prism block with the OP sample is illuminated monochromatically. We found some differences in the spectral bandwidths and maxima of the filters. Therefore, the amplitude transmittances of the interference filters have been measured with the aid of Varian Carry 5 VIS-NIR spectrophotometer and all deviations were considered in presentation of the refractometric results.

The right-hand collimator with the attached photo detector determines the measuring angle α (see Fig. 1). The angle of deviation γ is formed by the OP sample located into the *V*-shaped prism. The refractive index n_λ of the examined OP is calculated as follows:

$$n_\lambda^2 = N_\lambda^2 - \cos\gamma\sqrt{N_\lambda^2 - \cos^2\gamma}, \quad \gamma = 90^\circ - \alpha, \quad (3.1)$$

where N_λ is the refractive index of the VoF3 prism, γ is the calculated angle of the deviated beam, and α is the measured angle on the G5 set-up. The index N_λ of the SF3 glass is determined by the data published in [8] at standard spectral wavelengths. A new OptiColor program involving

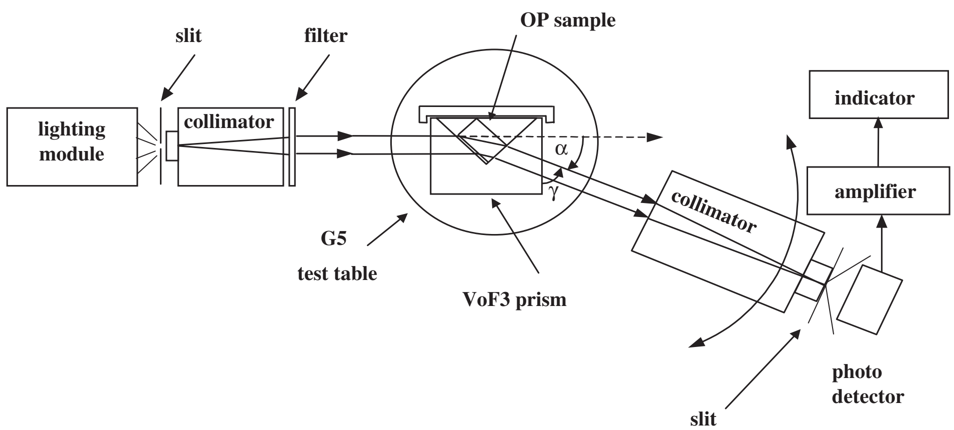

Table 1
Measured refractive indices of sixteen OP materials

Material	WLs (nm)					
	435.8	486.1	587.6	703	833	1052
PMMA	1.502	1.497	1.491	1.486	1.484	1.481
PS	1.617	1.606	1.592	1.582	1.577	1.572
PC	1.612	1.599	1.585	1.575	1.570	1.565
SAN	1.588	1.578	1.567	1.558	1.554	1.550
CTE Richardson®	1.602	1.593	1.580	1.571	1.566	1.562
NAS-21®	1.593	1.584	1.571	1.564	1.558	1.554
S (low styrene)®	1.532	1.526	1.518	1.512	1.509	1.506
Optorez 1330®	1.522	1.516	1.509	1.505	1.503	1.498
Zeonex E48R®	1.543	1.538	1.531	1.526	1.523	1.520
Bayer®	1.612	1.600	1.586	1.577	1.571	1.566
Cellulose[a]	1.480	1.477	1.471	1.466	1.463	1.461
Polyacrylate[a]	1.507	1.500	1.494	1.491	1.489	1.486
Styrene[a]	1.534	1.527	1.519	1.513	1.510	1.507
Polycarbonate[a]	1.597	1.587	1.572	1.565	1.560	1.555
Polystyrene[a]	1.615	1.604	1.592	1.582	1.576	1.572
Acrylic[a]	1.502	1.498	1.492	1.488	1.485	1.483

[a] Control samples.

Caushy's dispersion formula was made to determine the dispersive coefficients of SF3 glass prism and then random refractive indices N_λ in VIS or NIR region are calculated.

The obtained measured data of the refractive indices of OPs at six wavelengths are presented in Table 1. The last six materials from Cellulose to Acrylic refer to laboratory specimens and are used as control samples. The presented values of measured refractive indices in Table 1 were obtained by averaging over all measured data of each series of at least five samples at given wavelength.

4. Computer modelling of dispersion

In spectral regions where materials are transparent and normal dispersion occurs ($\lambda \gg \lambda_{0i}$) Eq. (2.4) is reduced to Cauchy's equation. In our previous works [7,12,25] we have applied a modified Cauchy's approximation in the form:

$$n_\lambda^2 = A_1 + A_2\lambda^2 + \frac{A_3}{\lambda^2} + \frac{A_4}{\lambda^4} + \cdots, \qquad (4.1)$$

where $A_1, A_2, A_3, A_4, \ldots$ are the calculated dispersion coefficients and λ is the wavelength expressed in microns.

We have studied the precision of this approximation in respect to the number of the involved dispersion coefficients. They were computed with the aid of linear systems consisting of four, five, six, seven and eight equations. The available optical catalogues do not provide sufficient data for refractive indices of OPs. Because of that, we have studied the accuracy of approximation (4.1) using the extensive refractometric information on optical glasses published in Glass Catalogues. Our calculations were made on the examples of catalogue data of SF3 [8] and SF6 glass [26]. The refractive indices are given with precision of 1×10^{-5}. Table 2 presents the results of our calculations for the SF6 type glass.

In column 2 the catalogue refractive indices at standard spectral lines are included. Columns 3, 4, 5, 6 and 7 present the deviation ΔN_λ between calculated and catalogue data. Blank places in these columns indicate the refractive indices used in the corresponding linear equation system involving 4, 5, 6, 7 or 8 dispersion coefficients in Eq. (4.1), respectively. A maximal error ΔN_λ of 0.00064 and 0.00026 in the blue area of the spectrum are obtained applying four and five dispersion coefficients. We found that the accuracy to the fifth decimal place, as in SCHOTT catalogue applying the Sellmeier's approximation, is not achievable. Using six dispersion coefficients a maximal deviation up to 0.00007 is obtained (column 5). One can see that utilization of more than six terms in the approximating row does not change significantly this result (columns 6 and 7). Therefore, we found that the usage of six dispersion coefficients in Cauchy's approximation is sufficient to provide the accuracy of ± 0.001 of calculated refractive indices. Better precision is achievable using Sellmeier's approximation, which should be applied in case of experimental data presented to the fourth decimal point.

Table 2
Deviation of computed indices in respect to the catalogue data of SF6 glass

WLs (nm)	SF6 (SCHOTT)	Calculated error ΔN_λ				
		4 coeff.	5 coeff.	6 coeff.	7 coeff.	8 coeff.
404.7 (*h*)	1.86436	0.00064	0.00026	−0.00005	−0.00023	−0.00020
435.8 (*g*)	1.84707					
480 (F')	1.82970	−0.00013	−0.00003	0.00001	0.00001	0.00001
486.1 (*F*)	1.82775	−0.00014	−0.00004			
546.1 (*e*)	1.81265	−0.00004	−0.00001	0	0	
587.6 (*d*)	1.80518					
589.3 (*D*)	1.80491	0	0	0	0	0
632.8 (laser)	1.79884	0.00002	0	0	0	0
643.9 (C')	1.79750	0	−0.00001	−0.00001	−0.00001	−0.00001
656.3 (*C*)	1.79609	0.00001				
706.5 (*r*)	1.79117					
852.1 (*s*)	1.78157	−0.00004	0	−0.00001		
1014 (*t*)	1.77517					

According to the analysis of the approximation precision best results are obtained using six dispersion coefficients in Cauchy's formula (4.1). We have realized the program OptiColor that allows us to compute the dispersion coefficients of any optical material in the region of normal dispersion. The input data consists of six refractive indices at selected measuring wavelengths (see Table 1). The dispersion coefficients from A_1 to A_6 can be calculated using the linear system consisting of six equations [7].

The obtained results are presented in Table 3. The computed dispersion coefficients vary in respect to the selected wavelengths, but analysis shows that A_1 always corresponds to n^2 with accuracy to the first decimal place and $A_2 \ldots A_6$ introduce additional corrections of higher accuracy.

Substituting the obtained dispersion coefficients in Cauchy's dispersion formula (4.1), random refractive indices can be computed and compared with their measured values. We have calculated OPs' refractive indices at selected laser wavelengths in our earlier works [7,12,27]. In this paper we present results at some additional laser emission wavelengths. The calculated refractive data of various OPs, SF3 and SF6 glasses are given in Table 4. The Abbe

Table 3
Computed dispersion coefficients of the examined OP materials

Material	Dispersion coefficients					
	A_1	A_2	A_3	A_4	A_5	A_6
PMMA	2.399964	−8.308636E-2	−1.919569E-1	8.720608E-2	−1.666411E-2	1.169519E-3
PS	2.610025	−6.143673E-2	−1.312267E-1	6.865432E-2	−1.295968E-2	9.055861E-4
PC	2.633127	−7.937823E-2	−1.734506E-1	8.609268E-2	−1.617892E-2	1.128933E-3
SAN	2.595568	−6.848245E-2	−1.459074E-1	7.329172E-2	−1.372433E-2	9.426682E-4
CTE Rich.®	2.663794	−1.059116E-1	−2.492271E-1	1.165541E-1	−2.211611E-2	1.545711E-3
NAS-21®	2.054612	1.374019E-1	3.200690E-1	−1.152867E-1	2.077225E-2	−1.383569E-3
S (low styrene)®	2.360004	−4.014429E-2	−8.371568E-2	4.160019E-2	−7.586052E-3	5.071533E-4
Optorez 1330®	2.291142	−3.311944E-2	−1.630099E-2	7.265983E-3	−6.806145E-4	1.960732E-5
Zeonex E48R®	2.482396	−6.959910E-2	−1.597726E-1	7.383333E-2	−1.398485E-2	9.728455E-4
Bayer®	2.542676	−4.366727E-2	−8.196872E-2	4.718432E-2	−8.892747E-3	6.324010E-4
Cellulose[a]	2.139790	−6.317682E-3	−5.920813E-3	9.613514E-3	−1.967293E-3	1.363793E-4
Polyacrylate[a]	2.364830	−6.955268E-2	−1.356107E-1	6.053900E-2	−1.166640E-2	8.542615E-4
Styrene[a]	2.274658	−5.700326E-3	−7.262838E-3	1.233343E-2	−2.481307E-3	1.784805E-4
Polycarbonate[a]	2.496875	−5.014035E-2	−4.188992E-2	1.732175E-2	−1.240544E-3	−1.977750E-5
Polystyrene[a]	2.721609	−9.982812E-2	−2.518650E-1	1.269202E-1	−2.549211E-2	1.867696E-3
Acrylic[a]	1.866120	2.085454E-1	4.806770E-1	−1.840693E-1	3.424849E-2	−2.340796E-3

[a] Control samples.

Table 4
Abbe numbers and refractive indices of two glasses and sixteen OP materials calculated for ten laser wavelengths

Optical material	Abbe numbers		Lasing medium									
			GaN	Ar	Cu	Nd:YAG	He–Ne	Ruby	Krypton	Ti:Sapphire	Nd:YAG	Nd:YAG
	ν_d	ν_{804}	405 nm	488 nm	510.6 nm	532 nm	632.8 nm	694.3 nm	799.3 nm	860 nm	946 nm	1064 nm
SF3	28.14	48.48	1.7879	1.7582	1.7530	1.7488	1.7347	1.7292	1.7226	1.7189	1.7166	1.7133
SF6	25.44	44.81	1.8641	1.8272	1.8209	1.8158	1.7988	1.7923	1.7845	1.7801	1.7775	1.7737
PMMA	59.2	96.9	1.516	1.497	1.496	1.495	1.489	1.487	1.484	1.484	1.483	1.481
PS	30.5	56.6	1.634	1.605	1.602	1.599	1.587	1.583	1.578	1.576	1.574	1.572
PC	29.1	54.8	1.631	1.599	1.595	1.592	1.580	1.575	1.571	1.569	1.567	1.564
SAN	35.4	66.8	1.608	1.578	1.575	1.573	1.563	1.558	1.554	1.553	1.552	1.549
CTE Rich.®	32.8	58.5	1.612	1.592	1.589	1.587	1.576	1.571	1.567	1.566	1.564	1.562
NAS-21®	35.5	56.3	1.588	1.583	1.580	1.577	1.568	1.564	1.559	1.557	1.555	1.554
S (low Styrene)®	44.9	79.6	1.542	1.526	1.524	1.522	1.515	1.512	1.510	1.509	1.508	1.506
Optorez 1330®	52.0	71.9	1.531	1.516	1.514	1.513	1.507	1.505	1.503	1.502	1.501	1.498
Zeonex E48R®	56.5	100.7	1.555	1.537	1.536	1.535	1.528	1.526	1.524	1.523	1.522	1.520
Bayer®	30.0	54.5	1.623	1.599	1.596	1.593	1.582	1.578	1.572	1.570	1.568	1.566
Cellulose[a]	54.1	84.3	1.484	1.476	1.475	1.474	1.469	1.467	1.464	1.463	1.462	1.461
Polyacrylate[a]	63.3	97.8	1.521	1.499	1.498	1.497	1.492	1.491	1.490	1.488	1.487	1.485
Styrene[a]	42.9	77.3	1.540	1.527	1.525	1.523	1.516	1.514	1.510	1.509	1.508	1.507
Polycarbonate[a]	28.9	56.7	1.602	1.586	1.582	1.579	1.568	1.565	1.561	1.559	1.557	1.554
Polystyrene[a]	32.0	55.5	1.640	1.604	1.601	1.598	1.587	1.582	1.577	1.576	1.574	1.571
Acrylic[a]	57.8	97.2	1.506	1.498	1.495	1.495	1.490	1.488	1.486	1.485	1.484	1.483

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