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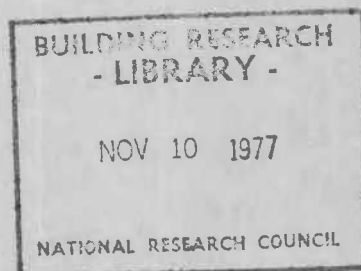
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ANALYZED

DISPERSION OF BIREFRINGENCE IN GLASSES AND ITS MEASUREMENT WITH THE BABINET COMPENSATOR USING WHITE LIGHT

by N.K. Sinha

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SOMMAIRE

L'analyse de l'utilisation du compensateur Babinet en conjonction avec la lumière blanche, pour la mesure de la contrainte provoquée à retardement relatif dans les verres, a révélé l'équivalence de la biréfringence spectrale normalisée du quartz cristallin et du coefficient de contrainte optique des verres ordinaires. On justifie l'utilisation de la lumière blanche sur une base limitée et on présente une brève description d'une formule courante de dispersion pour le quartz, le silice vitrifié et le verre calcosodique.

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Dispersion of birefringence in glasses and its measurement with the Babinet compensator using white light

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Analysis of the use of the Babinet compensator in conjunction with white light, for the measurement of stress induced relative retardation in glasses, revealed the equivalency of the normalised spectral birefringence of crystalline quartz and the stress optical coefficient of common glasses. Justification of the use of white light on a restricted basis and a brief description of a common dispersion formula for quartz, fused silica, and soda-lime-silica glass is presented.

There are many references to the occurrence of a black neutral fringe when Babinet wedges are used in white light to measure relative retardations in common glasses. This is usually explained by assuming negligible variation of the stress optical coefficient of glasses with wavelength. The validity of this explanation was questioned by the author when he observed a coloured neutral fringe (half bluish, half brown) while measuring the natural birefringence of single crystals of ice which is known to have negligible dependence on frequency in the visible range.

Basic photoelastic relations governing the operation of this compensator are reviewed and analysed in an effort to remove the discrepancy between the explanation and the observation. The analysis resulted in the proposal of a new method of presenting the dispersion of the stress optical coefficient of common glasses, which could conveniently be used to examine the inter-relationship between the optical responses of ordinary glasses and their parent member.

Photoelastic relation

The stress induced birefringence in a glass plate at room temperature is linearly related to the stress components by Brewster's law,

$$R = l(n_1 - n_2) = C_\sigma l(\sigma_1 - \sigma_2) \quad (1)$$

$$R = R(\lambda), C_\sigma = C_\sigma(\lambda)$$

where, R is the relative retardation, l the path length, and n_1 and n_2 are refractive indices corresponding to the directions of the principal stresses σ_1 and σ_2 respectively. The stress optical coefficient is usually expressed in the unit of Brewsters ($10^{-12} \text{ N}^{-1} \text{ m}^2$) and R is measured in nanometres.

The Babinet compensator

This is one of the most accurate and simple instruments for determining relative retardation. Its operation and construction are described by Monack & Beeton.⁽²⁾ Using monochromatic light, the relative retardation, $R(=n\lambda)$, is given by

$$R = kD \quad (2)$$

where D is the distance travelled by the wedge corresponding to n number of wavelengths, λ , and k is a constant for the particular compensator.

When stressed glass is introduced, the neutral fringe is displaced according to the relative retardation induced by the glass. Since monochromatic light gives a series of interference lines, it is often difficult to detect the displacement of the neutral fringe. White light, however, gives a dark neutral fringe and a series of alternating colours of higher orders⁽¹⁾ and this simplifies the detection of the displaced neutral fringe. For this reason, white light is used universally when measuring birefringence in glasses and the following analysis establishes the criteria which the dispersion of the birefringence of a material must satisfy in order to produce a black neutral fringe in white light.

Analysis of the Babinet compensator technique

The fundamental relation giving the retardation due to quartz wedges is⁽²⁾

$$R = D(n_e - n_o) \tan \theta \quad (3)$$

where $(n_e - n_o)$ is the birefringence of quartz, often denoted by β , and θ is the wedge angle. Comparing Equation (2) with Equation (3) gives

$$k = (n_e - n_o) \tan \theta = k(\lambda) \quad (4)$$

because the birefringence of quartz is dependent on wavelength⁽³⁻⁵⁾

$$(n_e - n_o) = (n_e - n_o)_\lambda \quad (5)$$

Thus in calibration, from Equation (2) and (4),

$$k(\lambda) = \frac{R(\lambda)}{D(\lambda)} \quad (6)$$

and in measurement on a specimen, from Equations (2) and (4),

$$R(\lambda) = k(\lambda)D(\lambda). \quad (7)$$

If λ_0 is a given standard wavelength and λ is any other wavelength, Equation (7) gives

$$\frac{R(\lambda)}{R(\lambda_0)} = \frac{k(\lambda)}{k(\lambda_0)} \cdot \frac{D(\lambda)}{D(\lambda_0)} \quad (8)$$

and Equation (1) gives

$$\frac{R(\lambda)}{R(\lambda_0)} = \frac{C_\sigma(\lambda)}{C_\sigma(\lambda_0)}.$$

From Equations (8) and (9)

$$\frac{R(\lambda)}{R(\lambda_0)} = \frac{C_\sigma(\lambda)}{C_\sigma(\lambda_0)} = \frac{k(\lambda)}{k(\lambda_0)} \cdot \frac{D(\lambda)}{D(\lambda_0)}. \quad (10)$$

Since D must be independent of λ , if the displaced neutral fringe is to remain black in white light, Equation (10) specifies the condition

$$\frac{R(\lambda)}{R(\lambda_0)} = \frac{C_\sigma(\lambda)}{C_\sigma(\lambda_0)} = \frac{k(\lambda)}{k(\lambda_0)} \quad (11)$$

or from Equations (4), (5), and (11),

$$\frac{R(\lambda)}{R(\lambda_0)} = \frac{C_\sigma(\lambda)}{C_\sigma(\lambda_0)} = \frac{(n_e - n_o)_\lambda}{(n_e - n_o)_{\lambda_0}} = r_\lambda \quad (12)$$

The ratios r_λ will be called the 'normalised' values and λ_0 will be called the normalising wavelength.

Isard & Douglas⁽⁶⁾ used white light for their stress relaxation experiments on silica; Van Zee & Noritake⁽⁷⁾ used white light for their well known investigation of the stress optical coefficient of plate glass at elevated temperature; Nordberg *et al.*⁽⁸⁾ demonstrated the stress profile in a glass plate after ion exchange using white light and quartz wedges; and Gardon and his group⁽⁹⁾ developed an automatic stress analyser using white light and Babinet wedges. There are innumerable examples, like these, of the use of white light with quartz wedges. It has been the common experience that the neutral fringe produced by white light was black for ordinary glasses even in its displaced position. Moreover, the fringe displacement measured with the white light was observed to be equal to the shift measured with monochromatic radiation. These observations must result in the equality given by Equation (12) which contradicts the assumption made by various authors that the stress optical coefficient, and hence the birefringence of ordinary glasses does not vary with the wavelength or, if it does, the variation is so small that it produces a negligible difference between the fringe displacement measured with white light and monochromatic radiation. The validity of Equation (12), however, could be examined only if the stress optical coefficient of common glass were known for the entire visible range.

Stress optical coefficient of plate glass

The spectral stress optical coefficient of labora-

tory annealed commercial soda-lime-silica glass has been determined by Sinha⁽¹⁰⁾ at room temperature in the 438–1150 nm radiation band and is presented in Table 1 together with the normalised quantities for a normalising wavelength of 546.1 nm. The reported results cover not only the visible range but a section of the near infrared.

Table 1. Stress optical coefficient of plate glass at 20°C and its normalised value as a function of wavelength; normalising wavelength 546.1 nm

Wavelength (nm)	$C_\sigma(\lambda)$ ($10^{-12} \text{ N}^{-1} \text{ m}^2$)	r_λ $C_{\sigma(\lambda)}/C_{\sigma(\lambda_0)}$
438.5	2.527	1.049
480	2.444	1.014
508.5	2.423	1.005
524	2.430	1.008
546.1	2.410	1.000
589.3	2.372	0.984
605	2.412	1.001
627	2.399	0.995
643.8	2.390	0.992
672.5	2.363	0.980
789	2.340	0.971
922	2.309	0.958
1000	2.309	0.958
1070	2.299	0.954
1150	2.299	0.954

The experiments were performed on a lapped and polished glass specimen approximately $12 \times 25 \times 150$ mm taken from a commercially annealed large plate. After lapping and polishing, the specimen was properly annealed in the laboratory using a controlled procedure. The annealed specimen was placed in a loading system which produced pure bending in its central part. Observations were performed, for a number of load levels, with collimated monochromatic light intersecting the specimen perpendicular to the length and parallel to the side faces. Two different ways of loading were used; light travelled either parallel or perpendicular to the original plate. In the former case, the total retardation was about twice as high (the optical path being about double) as in the latter case, but the error of measurement was larger due to the layered inhomogeneity. In each case, photographic recordings as well as manual measurements were made. An infrared image converter was used for visual measurements in the near infrared. A high pressure mercury lamp, a 40 W zirconium lamp, and spectral lamps were used as radiation sources. Very narrow bands of radiation were produced by high quality narrow band pass interference filters.

The compensator was calibrated using Equation (6) and the results verified for a few standard wavelengths by comparing the relative slopes $k(\lambda)/k(\lambda_0)$ either with the corresponding β/β_0 or with the values calculated using Equation (3) and the birefringence of quartz⁽³⁾ and the wedge angle determined by conventional spectroscopic methods. Variations of the slope of the calibration lines with wavelength for a few standard spectral lines are shown in Figure 1; experimental points corresponding to $n\lambda/2$ fringes are not shown for clarity. The line for 274.9 nm was calculated to illustrate the rapid increase in the slope

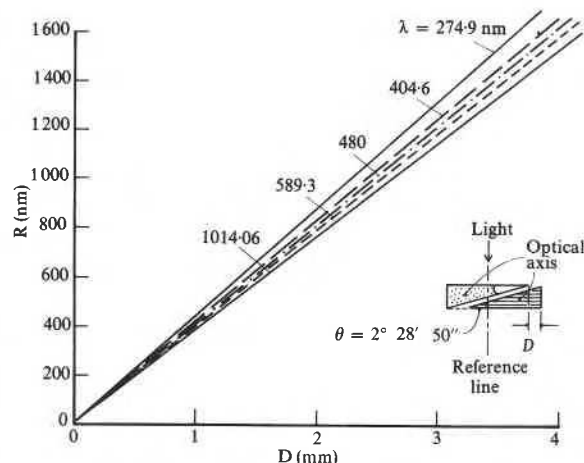


Figure 1. Calibration of the Babinet compensator, used in the present investigation

expected towards the shorter wavelength. A detailed description of the influence of the specimen and the optical system in shifting the nominal peak wavelengths and the determination of unknown 'dominant' wavelengths using Babinet compensator calibration data have been given elsewhere.⁽¹¹⁾

Both tensional and compressional birefringence were observed to vary linearly with stress up to 480 kg/cm² ($470.7 \times 10^5 \text{ N m}^{-2}$). The stress optical coefficient was determined from the experimental data, for various wavelengths, by the generally accepted statistical method. Highest measured relative retardations were approximately 3000 or 1500 nm for the maximum load of 480 kg/cm², depending on whether the light travelled parallel or perpendicular to the plate. In view of the well known fact that Babinet settings can only be made to about 1/50 fringe, or about 10 nm, the minimum error of measurements was about 3 and 6 in 1000 for the two cases. The uncertainty of measurement in the former was actually somewhat larger because of the striations in the glass. Results reported in Table 1 are considered to be accurate to about 0.5%. In the light of the error of measurements, the values of C_σ in Table 1 could be rounded off to two decimal places, but four significant (three decimal places) figures are maintained so that the normalised values are representative to three significant figures. Variation in the second decimal place has been observed by the author, however, for specimens from different batches of apparently the same composition. This could be due to slight variation of the commercial glass composition, water content, ageing, or previous thermal history. Even though the magnitude of the stress optical coefficient was slightly different for specimens from different batches, the normalised values were found to agree with each other within the experimental error.

The normalised values of the spectral stress optical coefficient of plate glass is compared in Figure 2 with the normalised spectral birefringence of quartz, the normalising wavelength being 546.1 nm. It can be seen that the prediction expressed by Equation

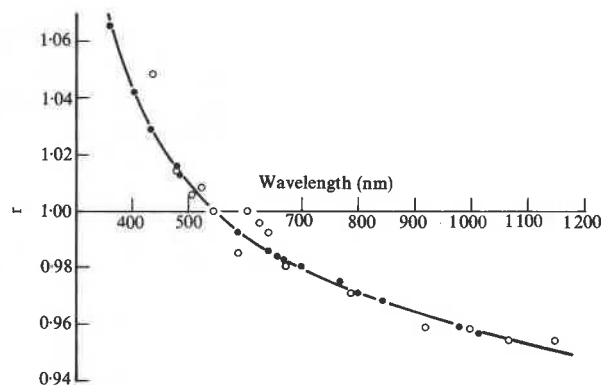


Figure 2. Comparison of the normalised stress optical coefficient of plate glass with the birefringence of quartz at 20°C; normalising wavelength 546.1 nm

— values calculated from Equation (13)
● birefringence of quartz
○ stress birefringence of plate glass

(12) is in agreement with the observations. That the colour of the neutral fringe in its displaced position is black in white during measurement of the stress induced birefringence of ordinary glass, is definitely a coincidence arising out of the equivalency of the normalised double refraction in both glass and quartz throughout the visible range.

It was mentioned earlier that the neutral fringe for ice was not black in its displaced position with white light. This can be considered now on the basis of the normalised birefringence of ice with respect to that of quartz. The normalised birefringences of ice are 1.071 and 1.000 at 404 and 706 nm respectively,⁽¹²⁾ whereas the corresponding normalised quantities for quartz are 1.042 and 0.979, the normalising wavelength again being 546.1 nm. These values for the two materials are considerably different from each other and therefore Equation (12) is not satisfied.

It would be of interest to see the colour of the neutral fringe for a high lead glass,⁽¹³⁾ for which, counter to the response of common glasses, the numerical value of the stress optical coefficient decreases towards the ultraviolet.

In one interesting case a slightly tinted neutral fringe was produced with white light for a thermally tempered glass plate (6 × 25 × 150 mm). The coloration was not obvious and it was noticed only when the colour of the neutral fringe was compared with that produced in a similar, but annealed, glass plate mechanically stressed to the same level and observed with the same source of white light. A possible explanation was found when the relative retardation in the tempered specimen was measured as a function of wavelength, normalised, and compared with the normalised spectral birefringence of the mechanically deformed specimen. This is shown in Figure 3. The reason for such a difference in the dispersion of a tempered glass was not known. Probably it was due to a certain 'freezing in' of high temperature structures during tempering. Further critical observations are necessary to give a valid explanation of this phenomenon.

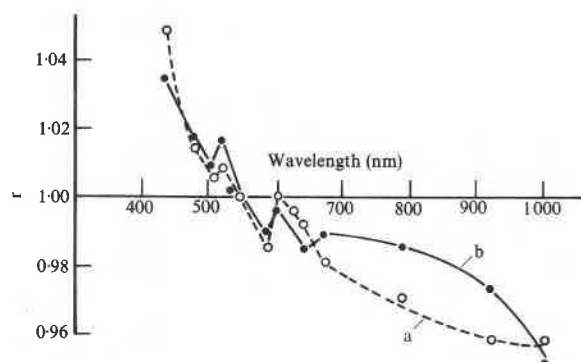


Figure 3. Normalised birefringence ($r_\lambda = R_\lambda/R_{\lambda 0}$) produced in a mechanically deformed specimen (curve a) compared with that in the mid-plane of a tempered specimen (curve b); normalising wavelength 546.1 nm. Relative retardations at the normalising wavelength were 1420 and 1540 nm for the mechanically loaded and the tempered specimens respectively

Justification of the use of white light

A survey of literature indicates that there is a common belief that if the stress induced relative retardation is expressed in nanometers, then it is independent of wavelength. This misconception might be related to the observed independence of the Babinet fringe displacement with respect to wavelength in most glasses. Monack & Beeton⁽¹⁾ gave a comprehensive description of the Babinet compensator and the order of colours appearing with white light, but they took it for granted that the neutral fringe would stay black when strained glass was introduced between the polariser and the compensator. The various colours of higher orders were explained on the basis of the stress induced relative retardation expressed in nm without any consideration of the variation of this quantity with wavelength. In the classical work of Adams & Williamson,⁽¹⁴⁾ even high residual stresses were expressed in nm without giving the measuring wavelength. Balmforth & Holland⁽¹⁵⁾ systematically measured the stress optical coefficient of some optical glasses and a series of soda-lime-silica glasses. They recognised the dependence of C_σ upon wavelength, but stated that over the range 546.1–589.3 nm, the stress optical coefficient was constant. The slight variation in the calibration of their Babinet compensator for the two wavelengths was apparently considered to be due to the uncertainty of the measurement. This was recognised by Van Zee & Noritake,⁽⁷⁾ but since they found that the neutral fringe displacement measured with monochromatic light gave the same value as with white light, they continued using white light. Gardon *et al.*,⁽⁹⁾ however, neglected the spectral dependence of C_σ for glass and stated, 'The millimicron is used here as the unit of retardation, which—unlike the number of fringes—is independent of the wavelength of light used'. They also stated that there was no particular advantage to using monochromatic light.

In general, the measurement of the relative retardation should be performed with monochromatic light

for which the stress optical coefficient of the material is known. The identification of the neutral fringe could be made with white light and, once it is identified, its location with monochromatic light would pose no problem.

The use of white light with a Babinet compensator is justified only if the spectral dispersion of the stress optical coefficient, and hence the spectral relative retardation of the material under consideration, is compensated exactly by the spectral dispersion of natural birefringence of quartz according to Equation (12). Care should be taken, however, in calibrating the compensator with monochromatic light. The birefringence measured with white light using the calibration for a given wavelength should be taken as appropriate for the calibrating wavelength, and conversion to the actual stress should take into account the stress optical coefficient of the material in question corresponding to this wavelength. For soda-lime-silica plate glass it appears to be appropriate to use white light. This cannot be generalised for glasses of other compositions, however, especially for optical glasses containing lead oxide and other oxides of heavy metals, and for high polymers used extensively for model analysis.

Discussion

Attention has been drawn to the similarity between the spectral dispersion of the birefringence of natural quartz, which is highly uniform in structure and has a long range order in the arrangement of silicon and oxygen atoms, and the spectral dispersion of stress induced birefringence in plate glass, which is amorphous and without any long range order. Moreover, plate glass is made from a mixture of SiO_2 , Na_2O , CaO and other minor constituents whereas natural quartz consists of an ordered arrangement of Si and O atoms only. It was observed that this similarity could be extended to the normalised stress induced spectral birefringence of fused silica, which is chemically the same material as natural quartz but physically amorphous and similar to plate glass. Discussion of the similarity between plate glass and quartz was limited to the visible and part of the near infrared range. The data available for fused silica^(16–18) allowed the extension of this to the ultraviolet.

A critical analysis of this subject revealed that the dispersion of natural birefringence of quartz and that of the stress induced birefringence of plate glass and fused silica could be predicted from the spectral normalised refractive indices of the materials and two quasi-resonant frequencies, one in the vacuum ultraviolet and the other in the medium infrared. The relation is expressed by

$$r_\lambda = \frac{\beta_\lambda}{\beta_{\lambda_0}} (\text{quartz}) = \frac{C_\sigma(\lambda)}{C_\sigma(\lambda_0)} (\text{fused silica, plate glass})$$

$$= \frac{n_{\lambda_0}}{n_\lambda} \cdot \frac{\lambda^2}{\lambda_0^2} \cdot \frac{(\lambda_0^2 - \bar{\lambda}_1^2)}{(\lambda^2 - \bar{\lambda}_1^2)} \cdot \frac{(\lambda^2 - \bar{\lambda}_2^2)}{(\lambda_0^2 - \bar{\lambda}_2^2)} \quad (13)$$

where n is the refractive index of the material, and λ_1 (121.5 nm) and λ_2 (6900 nm) are the resonant frequencies in the vacuum ultraviolet and in the infrared respectively. In the foregoing relation, the refractive index for quartz to be used is that corresponding to the ordinary waves, the other quantities are as already defined.

The development of Equation (13) will be discussed

Table 2. Comparison of the measured and calculated values of the normalised spectral birefringence of quartz. The experimental data are taken from Hardy & Perrin⁽⁷⁾, Jenkin & White⁽⁸⁾, and Physics Handbook⁽⁹⁾. The calculated values are based on Equation (13)

Wavelength (nm)	Measured	Calculated
185.5	1.5453	1.5443
199.0	1.4253	1.4270
226.5	1.2835	1.2817
257.3	1.1897	1.1907
274.9	1.1559	1.1559
361.1	1.0654	1.0637
404.6	1.0425	1.0403
343.1	1.0284	1.0285
480.0	1.0163	1.0144
546.1	1.0000	1.0000
589.3	0.9934	0.9929
656.3	0.9836	0.9842
670.8	0.9826	0.9826
768.2	0.9749	0.9733
794.8	0.9716	0.9712
844.6	0.9684	0.9675
1014.1	0.9564	0.9565
1159.2	0.9476	0.9480
1307.0	0.9389	0.9395
1395.8	0.9324	0.9343
1479.2	0.9280	0.9293
1541.4	0.9258	0.9254
1681.5	0.9149	0.9165
1761.4	0.9084	0.9111
1945.7	0.8942	0.8980
2053.1	0.8920	0.8899

in a later paper. Normalised values of birefringence of quartz calculated from Equation (13) are compared in Figure 2 and Table 2 with the available data⁽³⁻⁵⁾ from the vacuum ultraviolet to the medium near infrared. Success in predicting the normalised birefringence, not only in the visible but also in the ultraviolet and infrared, led to the establishment of a dispersion formula similar to Equation (13) for the refractive indices of these materials in the same wide spectral range.⁽¹⁹⁾ It is concluded that the normalising method offers a powerful tool for comparing the natural birefringence of the crystalline form (quartz) of a glass former (SiO₂) with the stress induced birefringence of the amorphous state (vitreous silica) of the parent material and plate glasses of the same family.

Goodman⁽²⁰⁾ proposed a novel idea on the strained mixed cluster model of glass structure. He pointed

out how glasses of the same family have properties related to the polymorphic forms of the parent member. The normalising method of comparing the optical responses seems to support Goodman's idea. It would be interesting to see how the optical properties (and hence the dielectric response) of cristobalite and tridymite compare with the common normalised dispersion formula proposed and whether this be extended to other families of glasses?

Conclusions

The stress optical coefficient of an ordinary glass is a function of wavelength of light. The natural birefringence of quartz and the stress induced birefringence of ordinary soda-lime-silica glass can be presented by a common normalised dispersion formula. This is responsible for the formation of a black neutral fringe in its displaced position when a Babinet compensator is used to measure the mechanically induced relative retardation in ordinary glasses in conjunction with white light. This compensator, however, exhibits a coloured neutral fringe when the normalised dispersion of the birefringence of the material is 'not equal' to that of quartz.

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