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LINEAR BIREFRINGENCE DISPERSION OF UNIAXIAL INORGANIC CRYSTAL CALCITE ESTIMATED BASED ON CHANNELED SPECTRA

The main refractive indices of natural inorganic uniaxial crystal $CaCO_3$ are determined using an Abbe refractometer equipped with a polarizing filter for the visible range. A source of white light equipped with seven colored filters was used for this purpose. The linear birefringence of calcite was computed as the difference between the main refractive indices. In order to obtain more information about the dependence of calcite linear birefringence on light wavelength, the channeled spectra method was used. Computed linear birefringence for channeled spectra is a continuous function of the light wavelength. Based on the birefringence values, some compensatory layers working for different purposes were computed for the yellow radiation of a sodium lamp.

Keywords: calcite, uniaxial symmetry, multi-quarter wave and multi half-wave compensatory layers.

1. Introduction

The anisotropic materials (inorganic crystals, polymers, liquid crystals) are characterized by different physical properties in various directions and have a wide range of applications in industry, science and medicine. In optics, transparent anisotropic materials [1–3] are used to compensate the optical pathway, to obtain or change the light polarization state, to searching the internal forces produced in transparent flying devices, in vessels under the action of marine streams, or in mechanisms supporting heavy loads.

In the principal coordinate system, *Oabc*, each anisotropic transparent medium possesses three values of its refractive index. Anisotropic uniaxial media [2] are characterized by $n_a = n_b = n_o$ and $n_c = n_e$, called ordinary and extraordinary refractive indices, respectively. The principal refractive indices are measured by using linearly polarized radiation having the electric field intensity in the plane *aOb* (ordinary refractive index) and parallel to the *Oc* axis (extraordinary refractive index), respectively. The difference

$$\Delta n = n_e - n_o \tag{1}$$

is called the linear birefringence [1, 2]. The birefringence is a material parameter depending on the light wavelength.

The optical axis of an anisotropic uniaxial materials is the direction of light propagation which an change its polarization state. For any other light propagation direction in anisotropic layers, light continuously changes its polarization state, because its ordinary and extraordinary components propagate with different velocities.

When the three values of the refractive index are different, the anisotropic material is called biaxial [2], because it possesses two optical axes.

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Between anisotropic substances, the inorganic crystal called calcite is known to have high values of linear birefringence in the visible range.

Calcite ($CaCO_3$) is one of the most widely used materials in anisotropic optics due to its high birefringence, high transparency both in the visible and near-infrared (VIS–NIR) spectral regions, and excellent optical stability [4]. Its trigonal crystal structure, associated with the R-3c space group (Fig. 1), consists of alternating planes of Ca^{2+} ions and trigonal planar CO_3^{2-} groups, in which the central carbon atom is bonded to three oxygen atoms at angles of approximately 120° [5]. This architecture determines the orientation of the optical axis (the c-axis) and leads to pronounced dielectric anisotropy. The CO_3^{2-} planes are approximately perpendicular to the Oc-axis, while the Ca^{2+} ions occupy interstitial sites, contributing to mechanical stability and to the distribution of the internal electric field [4–7].

Calcite is a negative uniaxial crystal, characterized by the relation $n_e < n_o$, and high values of linear birefringence (approximately 0.172 at 589 nm) [1, 4]. This property enables the separation and precise control of orthogonally polarized components, and it is essential in the design of optical elements that exploit the phase difference between the ordinary (o) and extraordinary (e) rays. The Sellmeier-type dispersion relations can be employed to calculate the refractive indices as a function of wavelength [4, 8]. Calcite is widely used in optical path compensators, multi-order wave-plates, and Wood-type polarizers, applications that exploit the large contrast between the two refractive indices of the uniaxial medium corresponding to the ordinary and extraordinary propagation modes [4]. In optical instrumentation and polarized microscopy, calcite is preferred due to its stability and the possibility of fabricating plates with controlled crystallographic orientation. It is also used as a reference standard in the characterization of uniaxial media and in the determination of optical properties in the mid-infrared (MIR) spectral region for granular materials and geological and planetary applications [9].

From a physicochemical perspective, calcite has a density of 2.71 g/cm^3 and a Mohs hardness of 3, properties that enable its processing into stable optical plates with precise tolerances [5]. Its low solubility in water and susceptibility to acidic environments require protective measures during the handling of optical components [4]. Commercial data confirm the calcite optical transmittance values exceeding 90% in the visible range and strict thickness tolerances for components produced by suppliers such as Thorlabs and Edmund Optics [10, 11].

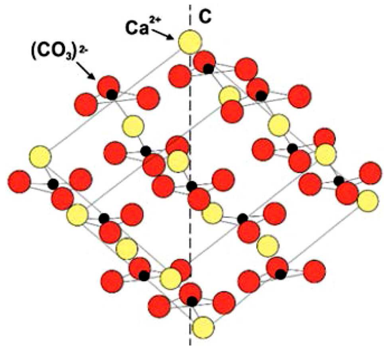


Fig. 1. Trigonal crystall structure of calcite ($CaCO_3$), illustrating the arrangement of Ca^{2+} ions and planar CO_3^{2-} groups along the crystallographic Oc-axis

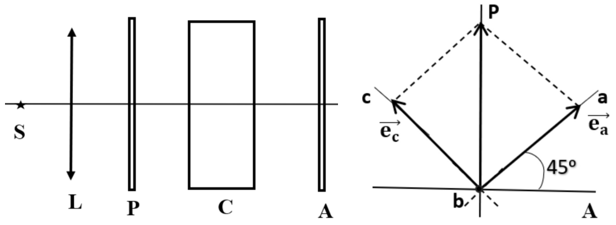
Due to different conditions of crystals growing in nature, there are differences in the transmittance or refractive indices values of calcite, so it is necessary to verify the applicability of the Sellmeier equations [4, 9] for a precise estimation of its principal refractive indices. The optical characterization of calcite involves the identification of its optical axis, the determination of its refractive indices n_o and n_e, and the estimation of the linear birefringence [9, 12].

The principal refractive indices of uniaxial crystals can be computed using known equations [4, 6, 9], but having into account that the growth of inorganic crystals depends on natural conditions [10], computed values may differ from the experimental data. The data used in designing the optical devices must be very precise [13] and experimental measurements are needed.

There are numerous optical methods [14, 15] for characterizing the birefringence of the anisotropic layers, such as interferometry [12, 16–18], conoscopy [19], ellipsometry [20, 21], method based on channeled spectra [22, 23], polarized microscopy [16], or refractometry in polarized light [23, 24] and so on.

These methods can be used for all transparent anisotropic uniaxial layers consisting of inorganic crystals [15–21], polymers [22–27], or liquid crystals [28–30].

The geometry of the refractive-index surfaces highlights the negative uniaxial nature of calcite: the



a *b*

Fig. 2. Device for obtaining the channeled spectra (S – source, L – lens, P and A – identical polarizing filters, C – inorganic crystal cut parallel to the optical axis) (*a*); relative orientation of the calcite principal axes *Oa* and *Oc* and the transmission directions of polarizing filters P and A (*b*)

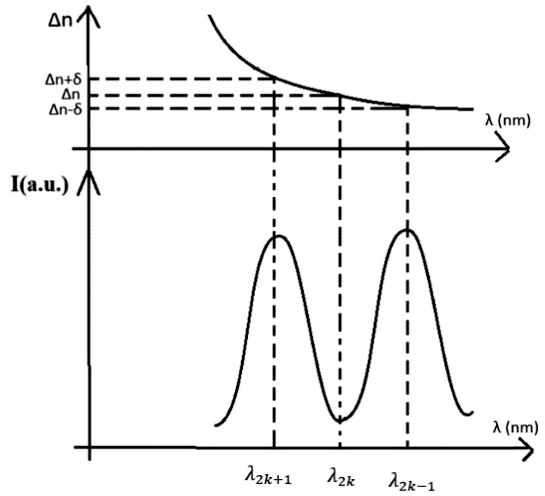


Fig. 3. Dispersive property of calcite and the part of channeled spectrum

ordinary refractive-index surface is spherical (n_o is constant in all directions), whereas the extraordinary refractive-index surface is an ellipsoid flattened along the c-axis, where n_e reaches its minimum value [1, 4]. This geometric representation allows the determination of propagation directions and phase velocities of the ordinary and extraordinary rays, providing a rigorous explanation of double refraction and beam separation in uniaxial crystals.

Through the combination of its distinct crystal structure, well-quantified optical properties, and physicochemical stability, calcite remains a benchmark material in anisotropic optics research and an indispensable component in applications requiring rigorous control of polarization, dispersion, and electromagnetic radiation propagation in uniaxial media [4–14].

2. Theoretical Notions

The method of channeled spectrum was used in order to determine the linear birefringence of calcite. The device used is schematically drawn in Fig. 2, *a*.

The source S is placed in the object focal plane of the lens L. The parallel beam propagates along the principal axis *Ob* of calcite. The axes *Oa* and *Oc* are contained in the entrance surface of inorganic crystal C which is introduced between the crossed polarizing filters P and A.

The orientation of the electric field of wave is determined by the transmission direction of the first polarizing filter P. Let us suppose that the azimuth angle between P and the principal axis *Oc* of crystalline layer is $\alpha = 45^\circ$. The components of the electric field on the principal axes *Oa* and *Oc* have equal magnitudes. The component along *Oa* is the ordinary component and propagates in crystal with velocity $v_o = c/n_o$ and the component parallel to *Oc*, called extraordinary, propagates with velocity $v_e = c/n_e$. In the propagation process a phase difference $\Delta\Psi$ is introduced between the two components:

$$\Delta\Psi = \frac{2\pi}{\lambda}\Delta n\, L, \tag{2}$$

Δn is the linear birefringence and L is the thickness of the crystalline layer in Eq. (2).

The monochromatic radiations satisfying the condition

$$\Delta\Psi = 2k\pi \tag{3}$$

at the exit from the crystalline layer are linearly polarized and have azimuth α (the same as at entrance in anisotropic layer). These light components cannot pass through analyzer A with its transmission direction perpendicular on P.

When radiations satisfy condition (4), the azimuth changes to $2\pi - \alpha$ and light passes through the analyzer A with the highest flux density

$$\Delta\Psi = (2k+1)\,\pi. \tag{4}$$

The other radiation components pass through the device at intermediary flux density. If the device D is introduced into a spectrophotometer, one obtains a channeled spectrum as a succession of maxima and null minima (when the azimuth angle satisfies $\alpha = 45^\circ$).

The linear birefringence of transparent anisotropic layer decreases as the light wavelength increases

(Fig. 3) and it influences the channels position. Let δ be the variation of the linear birefringence between successive components of light giving a minimum and its neighboring maxima.

Let us write the equations for the appearance of two maxima and the minimum between them:

$$\begin{cases} \dfrac{2\pi}{\lambda_{2k+1}}\left(\Delta n + \delta\right) L = (2k+1)\pi, \\ \dfrac{2\pi}{\lambda_{2k}} \Delta n L = 2k\pi, \\ \dfrac{2\pi}{\lambda_{2k-1}}\left(\Delta n - \delta\right) L = (2k-1)\pi. \end{cases} \tag{5}$$

By substituting the wavelengths λ with wavenumbers ν, and simplifying by π, one obtains:

$$\begin{cases} \nu_{2k+1}\left(\Delta n + \delta\right) L = \dfrac{1}{2L}(2k+1), \\ \nu_{2k}\, \Delta n\, L = \dfrac{2k}{2L}, \\ \nu_{2k-1}\left(\Delta n - \delta\right) = \dfrac{1}{2L}\,(2k-1). \end{cases} \tag{6}$$

The solutions of Eq. (6) can be written as Eq. (7)

$$\begin{cases} \Delta n = \dfrac{1}{2L}\dfrac{\nu_{2k+1} - \nu_{2k-1}}{2\nu_{2k+1}\nu_{2k-1} - \nu_{2k}(\nu_{2k-1} - \nu_{2k+1})}, \\ \delta = \dfrac{1}{2L}\dfrac{\nu_{2k-1} - \nu_{2k+1} - 2\nu_{2k}}{2\nu_{2k+1}\nu_{2k-1} - \nu_{2k}(\nu_{2k-1} - \nu_{2k+1})}, \\ k = \dfrac{1}{2}\dfrac{\nu_{2k}(\nu_{2k-1} - \nu_{2k+1})}{2\nu_{2k+1}\nu_{2k-1} - \nu_{2k}(\nu_{2k-1} - \nu_{2k+1})}. \end{cases} \tag{7}$$

From (7), it follows that the birefringence, Δn, the variation of birefringence, δ, between a channel and its neighbor and also the channel order, k, can be computed if the thickness of the crystalline layer and the wavenumbers of two maxima and the minimum between them are measured.

The advantage of channeled-spectra method is the rapidity; the values of the linear birefringence and its variation for all components of the visible range can be measured only from a single channeled spectrum.

3. Experimental Part

The first step in analyzing anisotropic materials is the establishing of principal directions in crystal [31, 32]. The optical axis of calcite (conventionally, Oc axis) was determined between crossed polarizing filters by rotating the crystal around the light propagation direction until the light did not pass through the system for any rotations. The other two principal axes [33] are perpendicular on the optical axis and have the characteristics that, if light propagates parallel to them, it does not change its polarization state. They were evidenced between crossed polarizing filters [1, 34].

Initially, some values of the principal refractive indices of calcite were determined with Abbe refractometer [23] at the monochromatic visible radiations using filters with known wavelengths.

In order to measure the principal refractive indices of calcite, [23, 24] two thin samples were cut from a calcite crystal, both parallel to optical axis, one parallel to aOc plane and the second parallel to bOc plane. They were introduced between the measuring prisms of Abbe refractometer equipped with polarizing filter. For each sample, the measurements were made for two orientations of the transmission direction of the polarizing filter (parallel to the principal axes of each sample). The ordinary refractive index, n_o, was measured for the transmission direction of the polarizing filter parallel to Oa, respectively Ob, while the extraordinary refractive index, n_e, was measured with transmission direction parallel to the optical axis Oc. The measurements were made using seven monochromatic filters (with known wavelengths).

In order to obtain information about the variation of linear birefringence within a small spectral range, the channeled spectra of calcite samples with two known thicknesses were recorded with a spectrophotometer Specord UV Vis Carl Zeiss Jena with data acquisition system. The linear birefringence and its variation were computed based on Eq. (7) using the values of neighboring maxima and minima obtained from the channeled spectra.

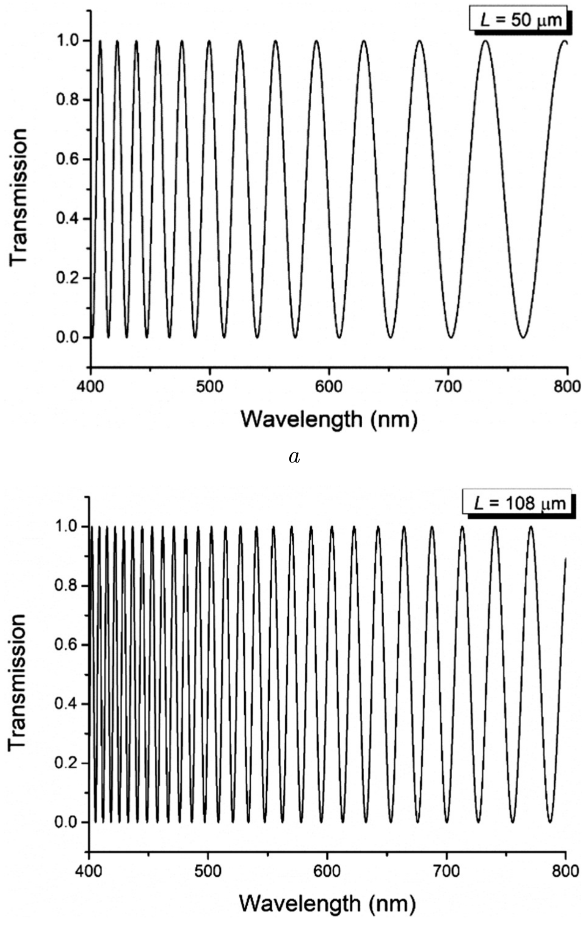
4. Results and Discussions

The obtained results in measuring the refractive indices using Abbe refractometer are given in Table 1, which also contains the values of the linear birefringence computed with Eq. (1).

The data from Table 1 show that calcite is a material with a high value of linear birefringence and with significant dispersion.

In order to increase the precision in determining the linear birefringence, we used the method of channeled spectra described in the theoretical section.

The channeled spectra of two calcite layers of 50 μm and 108 μm thicknesses are illustrated in Fig. 4.



a

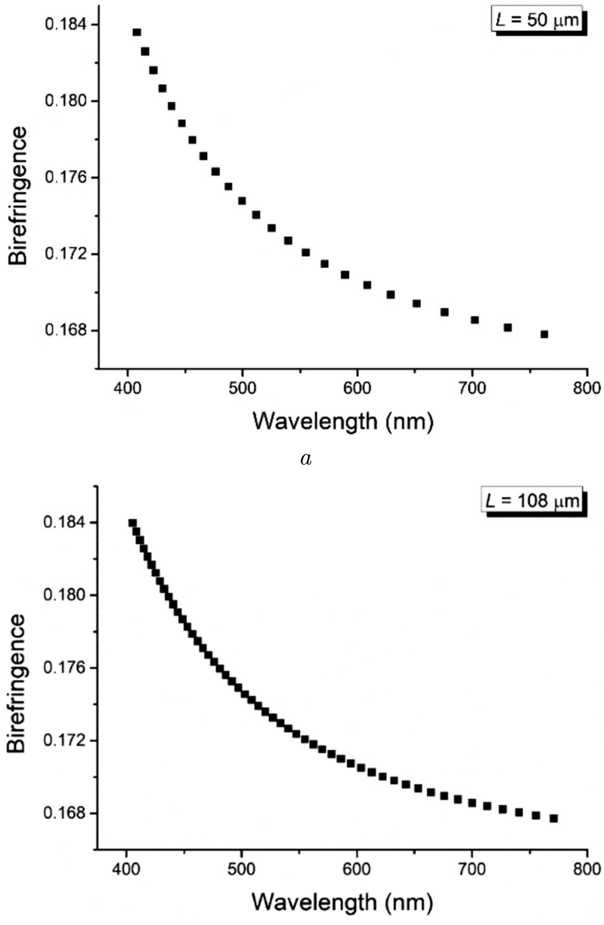
b

Fig. 4. Channeled spectra of calcite layers with thickness of 50 μm (*a*) and 108 μm (*b*), respectively

Table 1. **Principal refractive indices and linear birefringence of calcite**

λ (nm)	n_o	n_e	$\Delta n = n_e - n_o$
400	1.6782	1.4936	0.1846
450	1.6685	1.4892	0.1793
500	1.6619	1.4875	0.1744
550	1.6580	1.4862	0.1718
600	1.6555	1.4847	0.1708
650	1.6533	1.4840	0.1693
700	1.6521	1.4833	0.1688

As can be seen from Fig. 4, the number of channels increases with the increase in calcite sample thickness, contributing to the increase in measurement precision.



a

b

Fig. 5. Birefringence of calcite layers with thickness of 50 μm (*a*) and 108 μm (*b*), respectively

The wavelengths corresponding to the maxima and minima of the channeled spectra were measured and the thickness of the sample was known. The linear birefringence and its dispersion were computed based on Eq. (7). The linear birefringence of calcite in the visible range is illustrated in Fig. 5 for the two studied samples of calcite.

As can be seen from Fig. 5, the linear birefringence of calcite decreases with the light wavelength in the visible range, and the decrease is more accentuated in the range of short wavelengths.

The variation of the birefringence in the visible range is plotted in Fig. 6. This parameter increases when the light wavelength increases.

This study offers the possibility to increase the precision in determining the linear birefringence and its dispersion by increasing the thickness of the anisotropic layer.

5. Possible Applications of Calcite Layers Cut Parallel to Optical Axis

Due to its high value of linear birefringence, calcite is used as compensatory layers in various applications. The anisotropic compensatory layers must be computed for each monochromatic component of the visible light and the corresponding value of the birefringence. As anisotropic abundant material in nature, calcite is frequently used in optics, especially for layers with plan-parallel surfaces working as retarders.

The quarter-wave retarders transform the circular/elliptical polarized light into linear polarized light which is easily evidenced by a polarizing filter. This type of totally polarized light can be distinguished from the unpolarized light. The quarter-wave retarders are used in analyzing the polarization state of light. The thickness of quarter-wave retarder is computed with Eq. (8):

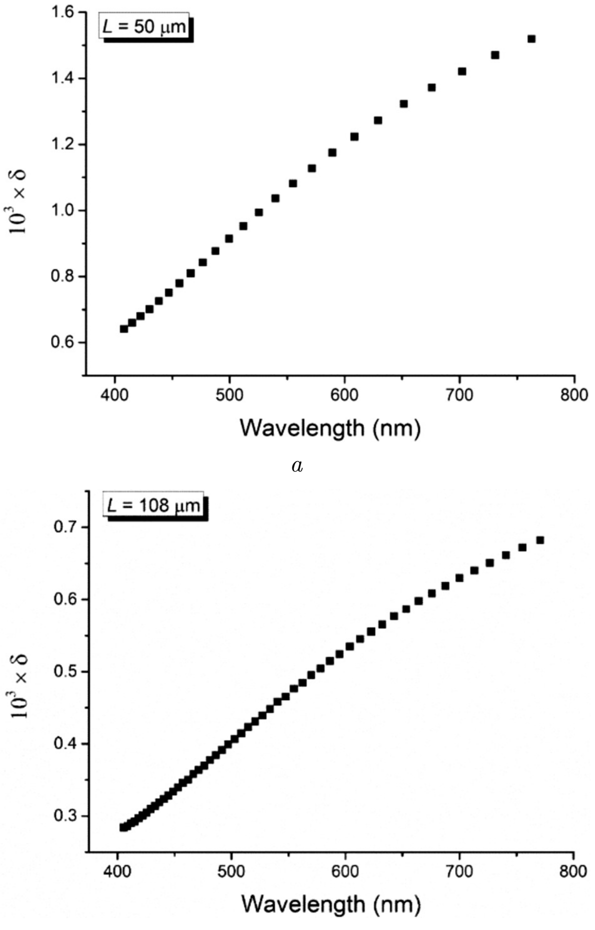
$$L_{\frac{\lambda}{4}} = \frac{(2k+1)\lambda}{4\,\Delta n}. \tag{8}$$

Some values of thicknesses of multi-quarter-wave layers are listed in Table 2. They are computed for high values of k, because calcite can be cut as thick layers.

The half-wave retarders are used in laboratories in which the optically active transparent materials are analyzed. The thickness of the half-wave retarders can be computed by Eq. (9):

$$L_{\frac{\lambda}{2}} = \frac{(2k+1)\lambda}{2\,\Delta n}. \tag{9}$$

This type of retarders changes a linearly polarized radiation with azimuth, α, into a linearly polarized radiation with azimuth $2\pi - \alpha$. When a half-wave compensator is introduced in the center of visual field of a polarimeter, two areas appear in the visual field in which the orientation of the electric field of light differs by 2π. In order to equalize the illuminations of the two areas from the visual field of the analyzer, its transmission direction must be the bisector of the angle 2α. When the active substance is introduced between the two polarizers, the rotation of the light electric field by an angle θ, determines a difference in illumination in the two areas of visual field due to



a

b

Fig. 6. Birefringence dispersion in the visible range for calcite layers with thickness of 50 μm (*a*) and 108 μm (*b*), respectively

Table 2. **Thickness of multi-quarter wave layers**

k	$L_{\frac{\lambda}{4}}$ (mm)	k	$L_{\frac{\lambda}{4}}$ (mm)
500	0.8565	2000	3.4269
750	1.2856	3000	5.1399
1000	1.7139	4000	6.8529

Table 3. **Thickness of multi-order half-wave layers**

k	$L_{\frac{\lambda}{2}}$ (mm)	k	$L_{\frac{\lambda}{2}}$ (mm)
500	1.7147	2000	6.8537
750	2.5172	3000	10.2797
1000	3.4277	4000	13.7040

different angles of the electric field in the two areas of the polarimeter visual field. The angle of the analyzer rotation around the light propagation direction until the visual field becomes uniformly illuminated evidences the rotation angle θ caused by the transparent optically active layer. The corresponding thicknesses of calcite layers for some values of k satisfying condition (9) for multi-half-wave retarders are listed in Table 3.

These layers are used to compensate the pathway introduced by various devices working in polarized light, or to measure the rotation angles introduced by the optically active transparent layers.

6. Conclusions

Calcite is a uniaxial inorganic crystal abundant in nature and characterized by high linear birefringence. Due to its resistance and low solubility in water, it is recommended for use in various devices for anisotropic optics.

The variation of the linear birefringence of calcite in the visible range is between 0.186 and 0.1688 in the wavelength range 400–700 nm. Our study contributed to determination of the birefringence and its dispersion for a wide range of closely spaced light components, due to the use of the channeled-spectra method.

Being transparent in visible range, the calcite crystal can be used as compensators of optical pathways. The crystal is cut parallel to a principal plane containing its optical axis and used at normal incidence.

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ОЦІНКА ЛІНІЙНОЇ ДИСПЕРСІЇ ДВОПРОМЕНЕЗАЛОМЛЕННЯ ОДНОВІСНОГО НЕОРГАНІЧНОГО КРИСТАЛА КАЛЬЦИТУ НА ОСНОВІ КАНАЛІЗОВАНИХ СПЕКТРІВ

За допомогою рефрактометра Аббе, оснащеного поляризаційним фільтром для видимого діапазону, визначено основні показники заломлення неорганічного одновісного природного кристала кальциту ($CaCO_3$). Для цього використовувалося джерело білого світла, оснащене сімома кольоровими фільтрами. Лінійне двопроменезаломлення кальциту обчислювалося як різниця між основними показниками заломлення. Для отримання додаткової інформації про залежність лінійного двопроменезаломлення кальциту від довжини хвилі світла використано метод каналізованих спектрів. Обчислене лінійне двопроменезаломлення для каналізованих спектрів є неперервною функцією довжини хвилі світла. На основі значень двопроменезаломлення розраховано деякі компенсаційні шари, що застосовуються в різних цілях, для жовтого випромінювання натрієвої лампи.

Ключові слова: кальцит, одновісна симетрія, багаточвертьхвильові й багатопівхвильові компенсаційні шари.