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The changes in the refractive index in optical glass, due to photothermal processing**

This work concerns the changes in refractive index within the visible range caused by the UV irradiation, the above problem being — to the authors knowledge — not represented in the literature. The preliminary results refer to the influence of the near ultraviolet radiation on the changes in refractive index in several glasses, irradiated at room and higher temperatures. The permanent local changes in the refractive index of order $\Delta n = 10^{-4}$ have been obtained without essential changes in optical absorption in the visible range. The UV irradiation of glass at suitably chosen temperatures seems to be the simplest method producing refractive index gradient in glass.

1. Introduction

An intensive examination carried out since some time has been aimed to obtain the glasses of prescribed refractive index gradient. The fused glass in subjected to a variety of actions in order to change its refractive index. Particulary great changes in the refractive index $\Delta n = 10^{-4}$–10^{-3} have been obtained in lithium–aluminium–silicon glasses and lithium–borium–silicon glasses by diffusion doping of sodium [1, 2, 3]. Sodium has been introduced to glass from the alloy containing the sodium ions by diffusion at higher temperatures. The doping with sodium causes the change in coordination of borium and aluminium [4].

This method of obtaining the glass with a refractive index gradient has some advantages (high value of Δn), and some disadvantages, of which the changes in the linear expansion coefficient of glass, accompanied with the changes in refractive index are most important. Consequently, some stress appearing during the glass cooling worsens the transparency of glass and induces the birefringence.

In the literature the influence at ionizing irradiation on the optical properties of glass is widely discussed. Among others it has been stated that due to irradiation of glasses by neutrons, γ-radiation or X-rays, the optical glass transmission worsens in the visible range, which is also accompanied by changes in refractive index. The magnitude of this effect depends upon the glass sort and the irradiation dose. The obtained values of refractive index were of order of 10^{-6}, whereby — depending on the glass composition — they were either positive or negative [5, 6]. These effects are considered to be disadvantageous, because they restrict the applicability of respective optical systems working under irradiation conditions. Therefore, some efforts have been undertaken to achieve such glass compositions, that would be insensitive to ionizing radiation. A possibility of practical application of the changes in refractive index occurring due to exposure of glass to ionizing radiation is limited by small values of Δn, on the one hand, and by the selective light absorption in the visible range, in the other. The changes in absorption occur due to creation and filling the electron trapes in the glass by ionizing radiation. The changes in refractive index are caused indirectly by the change in absorptivity of doping centres, and — according to the known Kramers–Kroning [7, 8] formulae — are proportional to the value of optical absorption coefficient. The results of the preliminary examinations have been presented below. Their purpose was to check the possibilities of obtaining the changes in refractive index of optical glass due to its UV-irradiation without simultaneous worsening of the optical transmission in the visible range.

2. The way of realizing the experiment

The glasses which were examined belonged to the crown glass group, like BK1, BK101, BK102, Bak104, containing tracing number of ions of changeable valency. As a light sources the following lamps were used:

1. High-pressure mercury HBO-200 lamp in housing of Zeiss make without additional filters.

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2. The Q 400 mercury lamp generating emission lines within the whole ultraviolet range.

3. Hydrogen laser of 337 nm of 0.5 MW power in one puls.

The glass samples of sizes $3 \times 10 \times 20$ mm cut out off the large block and polished were held at a given temperature within the 20–350°C range during irradiation. Half a sample has been irradiated the other half being protected by a metal plate, so that it was subjected to analogical thermal influence. The glass plates were also irradiated through a photographic film containing negative pictures. To measure the refractive index the Pulfrich PR-2 refractometer of Zeiss make were used. It should be noted, that results obtained in this measurement concern the value of the refractive index close to the sample surface. The magnitude of the refractive index change in the sample was estimated also with the help of Michelson type interferometer. The light source in this interferometer was the He–Ne laser ($\lambda = 633$ nm). The images irradiated through the photographic film were photographed in the optical system Δf shadowgraph without objectives. The chages in transmission were measured in spectrophotometer of Specord UV VIS type.

3. The results of measurements

As a measure of change in refractive index due to irradiation we have assumed the difference Δn between its values in the irradiated and nonirradiated regions; measured value of refractive index being characteristic of the near-surface layers.

The spectral dependence of changes Δn evoked by irradiation of different glasses under suitably chosen conditions has been presented in figs. 1–5. The same present also spectral dependence of optical transmission for these glasses in the nonirradiated (T) and irradiated (T_1) regions. The quantities T and T_1 have been expressed in percents without correction for refraction from the surface. Figs. 1 and 2 illustrate the changes occurring in two different glasses (BK7 —fig. 1, and BK101—fig. 2) due to an analogical photothermal processing, consisting in irradiation of the samples with an HBO-200 lamp at the temperature about 100°C for 1 hour. In fig. 3 the changes Δn are shown for two samples of the same BK107 glass irradiated at the temperatures 50°C (Δn_1) and 100°C (Δn_2), respectively. In fig. 4 the changes in Δn due to irradiation with the HBO-200 lamp has been presented as depending on the temperature at which the sample was held during illuminations. The measurement of the refractive index have been

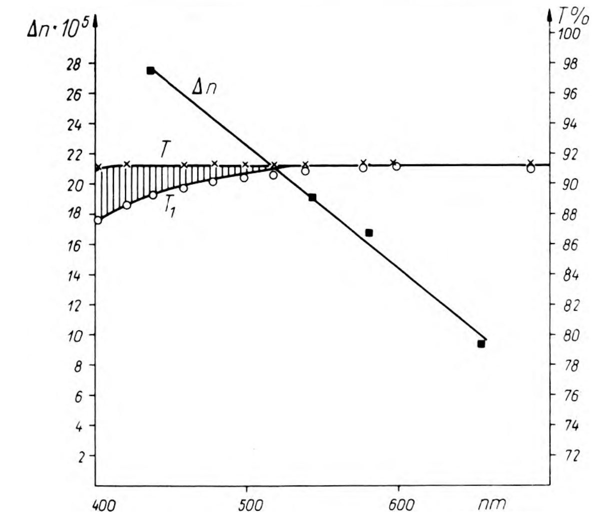

Fig. 1. The dependence of the refractive index changes (Δn) and optical transmission in BK7 glass, due to irradiation with the HBO-200 lamp at the sample temperature 100°C. The exposure time — 1 hour

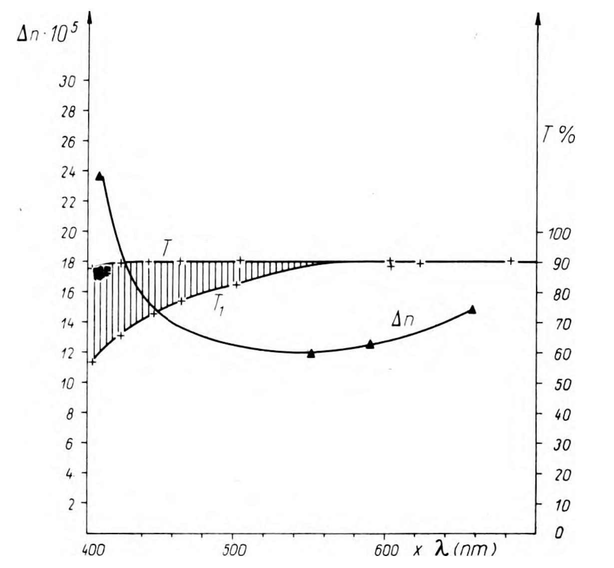

Fig. 2. The dependence of the refractive index changes and optical transmission in BK101 glass, due to irradiation with the HBO-200 lamp at the sample temperature 100°C. The exposure time — 1 hour

carried out for the mercury line ($\lambda = 546.1$ nm). As it is visible the curve reaches the maximum for the temperature of about 180°C, thus so at this temperature the maximal values of Δn is obtained.

The graphs presented in fig. 5 illustrate the dependence of Δn upon the irradiation time for two different illuminating sources. The first source was a mercury lamp with all the lines in the ultraviolet range (curve *2*), and the second source was a nitrogen laser of 337 nm wavelength and 0.5 MW power in single pulse (curve *1*). The samples were illuminated at room temperature. The measurements of Δn

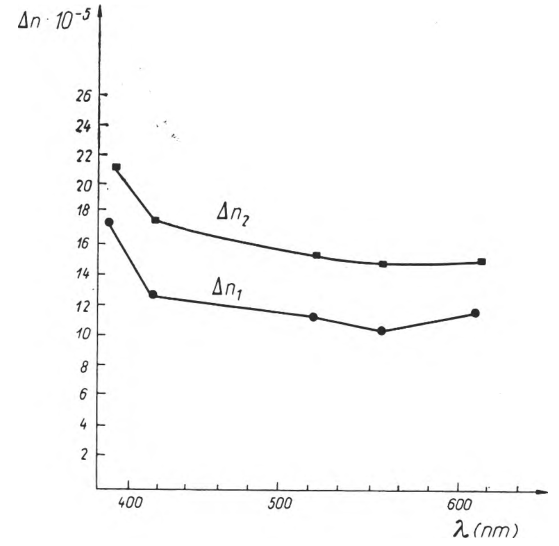

Fig. 3. The dependence of the refractive index changes (Δn), due to irradiation with the HBO-200 lamp at the sample temperature 50°C (Δn_1), and 100° C (Δn_2) in the BK107 glass. The exposure time — 1 hour

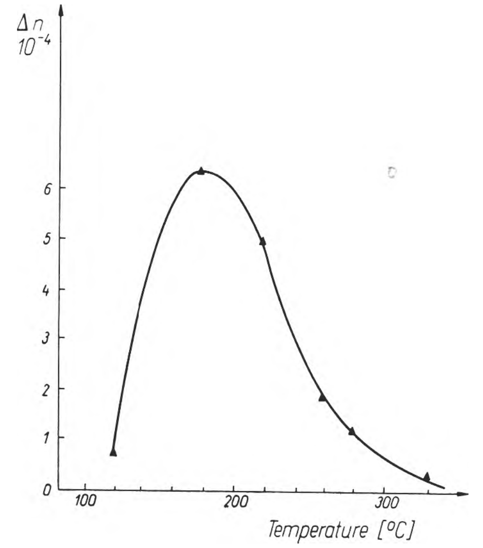

Fig. 4. The dependence in the refractive index changes (Δn) in BK107 glass upon the temperature of the sample during the irradiation. The exposure time — 1 hour. The measurements of refractive index were made for the line $d(\lambda = 587.6$ nm)

were carried out for the *d*-line ($\lambda = 587.6$ nm) As it may be seen the greatest changes in Δn are obtained in the course of first five minutes of irradiation.

The duralibity of the changes in refractive index has been also examined. The two interferograms presented in fig. 6 prove the stability of the changes. They were performed in the period of time of several weeks for the same sample of BK7 glass irradiated previously (in the pattern visible in the interferograms). As it can be seen no alteration at all happened in the shift of interference fringes.

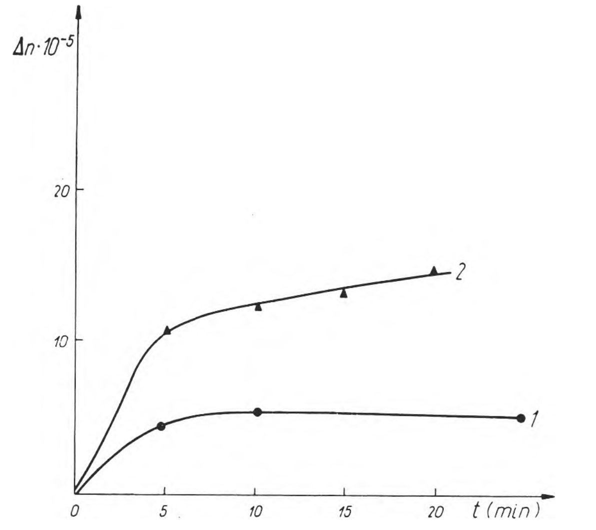

Fig. 5. The dependence of the refractive index changes (Δn) upon the irradiation time for BK101 glass. The irradiation at room temperature: a) with a mercury lamp (curve *2*), b) with a nitrogen laser, $\lambda = 337$ nm (curve *1*). The refractive index has been measured for $\lambda = 587.6$ nm

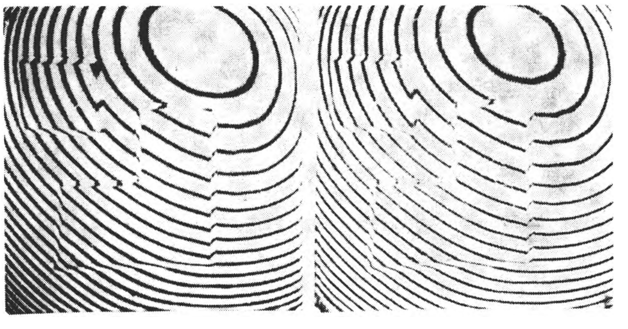

Fig. 6. The interferograms of the BK7 glass plate irradiated with a HBO-200 lamp at room temperature, a) made immediately after irradiation, b) made after a couple of weeks

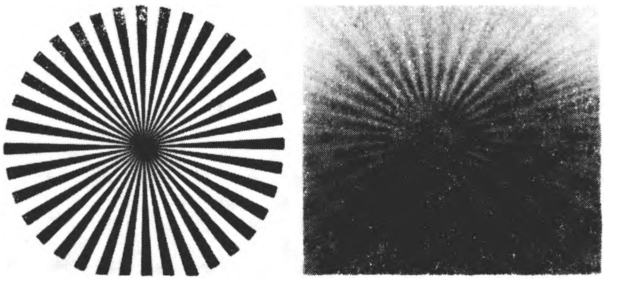

Fig. 7. The picture produced in BK101 plate by irradiation with the HBO-200 lamp at room temperature during 1 hour; a) photo of the test, b) effect of irradiation observed in the shadowgraph

In fig. 7a photograph of a test has been shown. A plate of BK101 glass of thickness 0.5 mm has been illuminated through the picture of this test fixed on the photographic film. Fig. 7b shows the photograph of glass, performed in this way in the shadowgraphs without objectives. This picture illustrates the refractive index gradient produced in this plate.

4. Discussion of results

It should be emphasized that the results obtained above are preliminary and that the purpose of this investigations was to recognize the problem, as this kind research seems to be unpublished so far in the literature. The magnitude of refractive index changes as well as their stability are interesting for practical applications.

The refractive index changes obtained so far have been of order of 10^{-5}–10^{-4}. This changes were durable and the control measurements made in several weeks periods gave the same results.

Although the changes in refractive index obtained by this method were very small but we were far from the optimal conditions so far as glass composition for this kind of interactions is concerned. The effectivity of interaction may be increased by the choosing more advantageous spectral range and dose of radiation. As the right temperature during the irradiation is an essential problem, there are some reasons to hope that this method may result in considerably higher changes in refractive index. The advantage of this method of producing the glass of variable refractive index lies in this simplicity and the fact that the irradiation conditions may be easily modified. The changes in optical absorption are small. Some BK101 glass samples may to high degree be thermally discoloured by annealing at 200°C without any influence on the distribution of the refractive index.

The examinations carried out so far are not yet sufficient to present the mechanisms of the process responsible for the change in the refractive index due to UV-irradiation. It is well known, however, that the change in refractive index should be connected with the change in the degree of ions polarization occurring due to oxidation and reduction processes.

Изменения коэффициента преломления в оптическом стекле под влиянием фототермической обработки

Работа касается коэффициента преломления стекла для видимого света в случае ультрафиолетового облучения. Этот вопрос в литературе пока не обсуждался. Представлены результаты предварительных исследований по влиянию излучения области, близкой к ультрафиолетовой, на перемены показателя преломления в нескольких стеклах, причем стекла облучались как при комнатной, так и при более высоких температурах. Получены устойчивые местные изменения коэффициента преломления порядка $\Delta n = 10^{-4}$ без существенных изменений в оптическом поглощении стекла в видимой области. Представляется, что ультрафиолетовое облучение при соответственно подобранной температуре является простейшим из применявшихся до сих пор методов создания градиента коэффициента преломления в стекле.

References

[1] Livshic V. J., Karapetyan G. O , Ilin V. G., Negodaev G. D., Fizika i Khimiya Stekla, Vol. 2, No. 1 p. 68, 1976.

[2] Livshic V. J., Karapetyan G. O., Negodaev G. D., Fizika i Khimiya Stekla, Vol. 3, No. 1, p. 28, 1977.

[3] Mazurina E. K., Jachkind A. K., Fizika i Khimiya Stekla, Vol. 2, No. 6, p. 551, 1976.

[4] Appen A. A., *Khimiya Stiekla*, Izd. Khimiya, Leningrad 1974.

[5] Olson D. R., Dieselman H. D., Schraeder I. B., Appl. Opt. Vol. 10, No. 1, p. 81, 1971.

[6] Byurganovskaya G. V., Kisin B. J., Orlov N. J., Optiko-Mekhan. Promyshlennost Vol. 34, No. 3, p. 34, 1967.

[7] Born M., Wolf E., *Principles of Optics*, Pergamon Press, London 1965.

[8] Kucharski M., Postępy Fizyki Vol. 27. No. 6, p. 597, 1976.

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