

Investigation of usability of the selected types of glass in planar waveguide technology

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Presented in the paper is a comparison of ion exchange $\text{Ag}^+ - \text{Na}^+$ through planar diffusion and electrodiffusion in certain types of glass in view of their utilization as substrates for single and multimode structures. The investigations included the determination of the magnitude of changes in the refractive index and rate of exchange, and the determination of the technological parameters of the distribution profile of the refractive index.

1. Introduction

One of the effects of carrying out ion exchange in glass is the change of the refractive index. It is a result of the combination of two principal phenomena: changes of the size of the exchanged ions and the difference of their polarizability [1]. This phenomenon is used for the production of waveguide structures in glass substrates [2]. The selection of a suitable substrate material is a problem to which insufficient attention has been paid in literature, although as shown by experience, the chemical composition of glass is decisive to a great extent about such important factors as the quantity of the changes in the refractive index, the rate of the diffusion process [3], [4], and the material attenuation.

$\text{Ag}^+ - \text{Na}^+$ ion exchange is, at present, the most common technology of producing the gradient waveguide structures [1]. Melted AgNO_3 [2] is most frequently used as the source of admixture in the temperature range 500-675 K.

The subject of this investigation is to find the characteristics of certain types of glass in view of their utilization as a substrate for single- and multimode structures. The research made refers to diffusion and electrodiffusion of Ag^+ ions and includes:

- testing of the rate of the process,
- determination of the magnitude of changes of the refractive index,
- determination of the profile distribution of the refractive index.

2. Theoretical description of the process of ion exchange

2.1. Ion exchange without external electric field

The basis of every ion exchange process is the phenomenon of two-component diffusion (Fig. 1). In this process, the substituted ions "a" of the concentration n_a take the place of the ions "b" of the concentration n_b , which are the natural

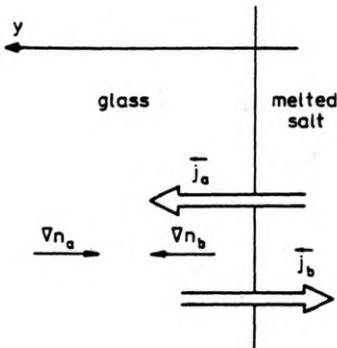


Fig. 1. Ion exchange without external electric field

components of the glass substrate (we assume that these are the only mobile substrate ions at the given temperature).

The differences of electric polarizability and the dimensions of the ion radius lead to the formation of the distribution of the refractive index proportional to the concentration of the n_a ions introduced. In an analysis of this process, the flux j_a of "a" type ions directed inside the medium, should be taken into consideration, as well as the flux j_b of "b" type ions, directed towards the surface:

$$\vec{j}_a = -D_a \nabla n_a + \mu_a \vec{E}_d n_a, \quad (1)$$

$$\vec{j}_b = -D_b \nabla n_b + \mu_b \vec{E}_d n_b \quad (2)$$

where: μ_a , D_a , μ_b , D_b denote the mobility and the coefficient of diffusion of "a" and "b" ions.

The intensity of the electric field E_d is related to the difference in the mobility of the exchanged ions. The electric field E_d accelerates the diffusion of slower ions and hampers the diffusion of faster ones, hence, it is a factor coupling the migration of both types of ions. Steady state is created at such a value of the field E_d at which the following equality takes place

$$\vec{j}_a = -\vec{j}_b. \quad (3)$$

Taking into consideration, in Eq. (1) or (2), the condition of continuity and the condition of electric neutrality the diffusion equation for the substituted ions is obtained

$$\frac{\partial n_a}{\partial t} = \text{div} \left[\frac{D_a n_0}{n_0 - (1-r)n_a} \text{grad}(n_a) \right] \quad (4)$$

where: n_0 - equilibrium concentration, $r = \mu_a/\mu_b$.

For one-dimensional diffusion (planar waveguide), in accordance with the denotations (Fig. 1), this equation is reduced to the form

$$\frac{\partial N_a}{\partial t} = \frac{\partial}{\partial y} \left[\frac{D_a}{1 - (1-r)N_a} \frac{\partial N_a}{\partial y} \right], \quad N_a = n_a/n_0. \quad (5)$$

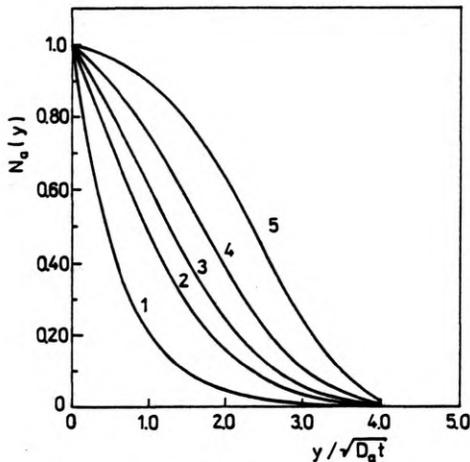


Fig. 2. Solution of the diffusion Eq. (5) for the different values of the parameter r (1 - $r=5$, 2 - $r=1$, 3 - $r=0.5$, 4 - $r=0.2$, 5 - $r=0.025$)

The sources of admixture used most frequently in practice in the form of melted salts of the suitable chemical compounds may be considered to be infinite sources. Hence, the boundary condition and the initial condition for Eq. (5) have the following form:

$$N_a(0, t) = 1, \quad N_a(y, 0) = 0. \quad (6)$$

The numerical solution of Eq. (5) with conditions (6) for the different values of r is presented in Fig. 2. Here, the very essential dependence of the character of solution on the value of this parameter should be noted.

2.2. Ion exchange in the presence of an external electric field

A process making possible the formation of deep profiles of changes in the index of refraction is the diffusion in the presence of an external constant electric field (electrodifusion). The external electric field E_0 forces a constant flow of ion current in the direction from the upper plane of the substrate to the lower one (Fig. 3).

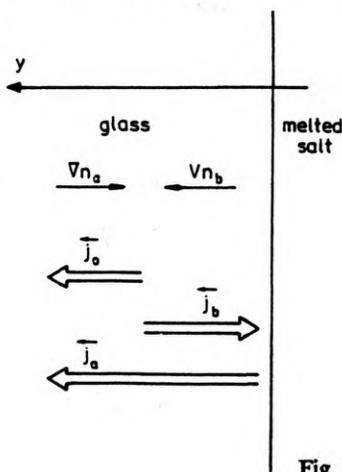


Fig. 3. Ion exchange with an external electric field

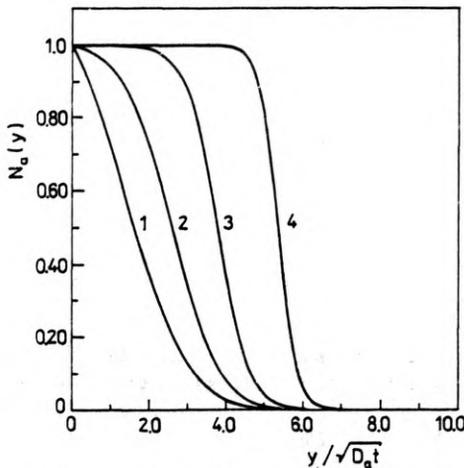


Fig. 4. Solution of the electrodiffusion Eq. (10) for the different values of the parameter r (1 - $r = 1$, 2 - $r = 0.5$, 3 - $r = 0.3$, 4 - $r = 0.2$)

The resultant flux of ions (current) is the sum of both fluxes of ions of type "a" and "b":

$$\begin{aligned} \vec{j}_a &= -D_a \text{grad}(n_a) + \mu_a (\vec{E}_d + \vec{E}_0) n_a, \\ \vec{j}_b &= -D_b \text{grad}(n_b) + \mu_b (\vec{E}_d + \vec{E}_0) n_b \end{aligned} \quad (7)$$

where: \vec{E}_d - internal electric field coupling the migration of the slower and faster ions, \vec{E}_0 - external electric field.

In a steady state

$$\vec{j}_a + \vec{j}_b = \mu_b n_0 \vec{E}_0 = \vec{j}_0 \quad (8)$$

where: \vec{j}_0 is the flux of substrate ions far from the glass surface. Making use of (8), we obtain an equation describing the process of electrodiffusion in the form

$$\frac{\partial N_a}{\partial t} = \text{div} \left[\frac{D_a}{1 - (1-r)N_a} \text{grad}(N_a) \right] - \mu_a \vec{E}_0 \text{grad} \left[\frac{N_a}{1 - (1-r)N_a} \right]. \quad (9)$$

This equation in the one-dimensional case (Fig. 3) is reduced to the form

$$\frac{\partial N_a}{\partial t} = \frac{\partial}{\partial y} \left[\frac{D_a}{1 - (1-r)N_a} \frac{\partial N_a}{\partial y} \right] - \mu_a E_0 \frac{\partial}{\partial y} \left[\frac{N_a}{1 - (1-r)N_a} \right] \quad (10)$$

where $N_a = n_a/n_0$.

The solution of this equation for the initial condition $N_a(y, 0) = 0$ and boundary condition $j_a(0) = \frac{D_a}{1 - (1-r)N_a} \frac{\partial N_a}{\partial y} = \mu_b E_0$ for $y = 0$, and the different parameters of r is presented in Fig. 4.

3. Experimental set-up

The processes of ion exchange without electric field (diffusion), as well as with an electric field (electrodiffusion) were carried out using pure silver nitrate AgNO_3 as a source of ions of the admixture (Ag^+). Process temperatures 573–623 K

were used. The intensities of the electric field E_0 assumed the values from the interval 0–150 V/mm. The temperature of the process was stabilized at 0.5% level. After carrying out the electrodiffusion processes, a special set-up (Fig. 5) was constructed, making possible the realization of the process in the flat glass plates. This eliminates the necessity of preparing special glass vessel, only fragments of which

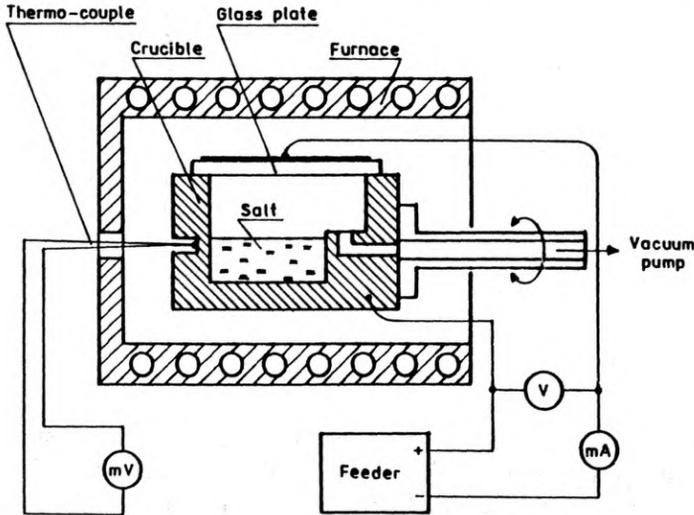


Fig. 5. System for the conducting of the processes of electrodiffusion

are used later. An advantage of this set-up is also a possibility of an accurate control of the duration of the process. The contact of the melted salt with the glass surface takes place after the turning of the crucible. This permits the avoiding of ambiguities of the duration of the process, associated with the heating from the moment of the melting of salt and reaching the proper temperature. The cathode was a layer of conducting glue superimposed on the other surface of the glass plate. The current flowing through the sample was controlled during the process of electrodiffusion.

4. Results of the experimental studies

The obtained distribution profiles of the refractive index for the processes of diffusion and electrodiffusion were determined by IWKB method on the basis of the measurement of the mode propagation constants ($\lambda = 0.6328 \mu\text{m}$). By fitting the theoretical curves which were the solutions of Eq. (5) to the experimental points (Figs. 6–9), the constants of the diffusion of ions Ag^+ were determined, as well as the ratio of their mobility r to the mobility of the surface ions Na^+ participating in the process of exchange.

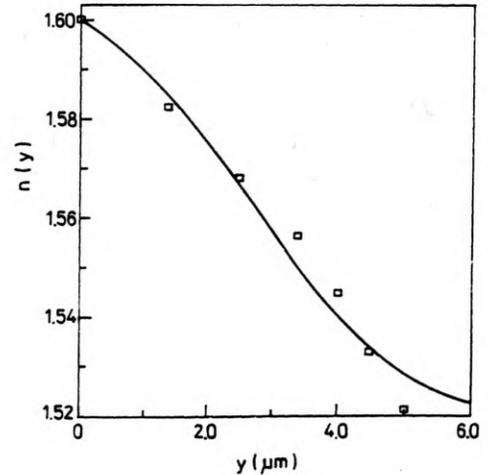
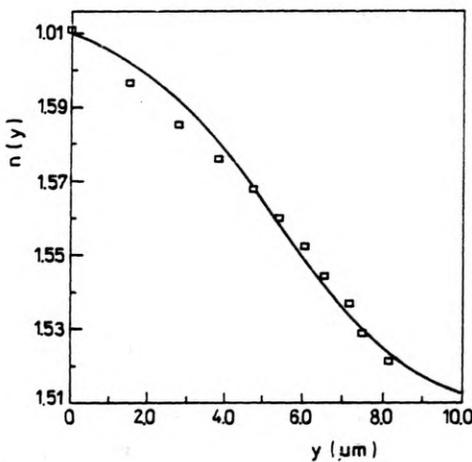
In type "A" glass, thanks to high sodium contents (Table 1), the process of diffusion takes place quickly, giving a significant change of the refractive index

Table 1. Chemical composition of investigated glasses

Component	Type of glass [% mol]			
	A	B	C	D
SiO ₂	75.11	69.58	80	60
Na ₂ O	12.21	8.44	3	15
B ₂ O ₃	—	9.91	12	14
CaO	7.43	0.07	0.2	—
MgO	3.9	0.07	—	—
BaO	—	2.54	1	—
K ₂ O	0.21	8.37	0.7	—
ZnO	—	—	—	8
Al ₂ O ₃	0.58	0.04	2	3
As ₂ O ₃	—	0.09	—	—
AsO ₃	—	0.22	—	—
Fe ₂ O ₃	0.01	—	—	—
<i>n</i>	1.511	1.520	1.471	1.511

(Data acc. to "Białystok" Glass Works)

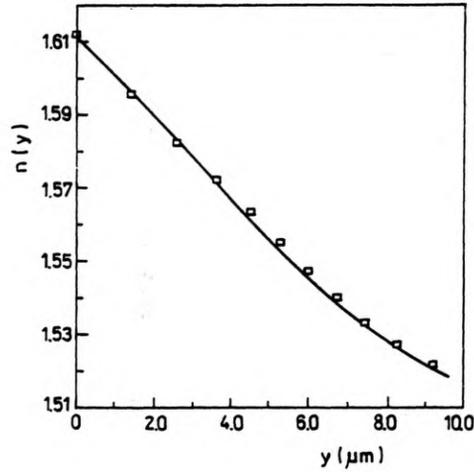
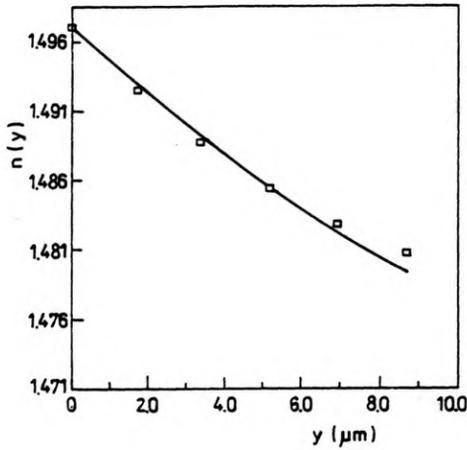
($\Delta n = 0.1$). However, the waveguide structures obtained in this type of glass are characterized by yellow dye penetration passing into yellow-brown one and exhibit significant material attenuation reaching 10 dB/cm. The occurrence of dye penetration in this type of glass is the result of reduction of Ag⁺ ions and their agglomeration into colour centres caused by the presence of iron and arsenic compounds [2] used to clarify the glass in the process of melting. A similar



▲

Fig. 6. Refractive index distribution in the glass of the type "A" after diffusion of Ag⁺ type ions ($T = 573$ K, $t = 1$ h). □ - experimental points determined by the IWKB method, solid line - numerical solution of Eq. (5)

Fig. 7. Same as in Fig. 6, but for "B" type of glass ($T = 573$ K, $t = 4$ h)



▲
Fig. 8. Refractive index distribution in the glass of the type "C" after diffusion of Ag^+ type ions ($T = 603 \text{ K}$, $t = 4 \text{ h}$). \square - experimental points determined by the IWKB method, solid line - numerical solution of Eq. (5)

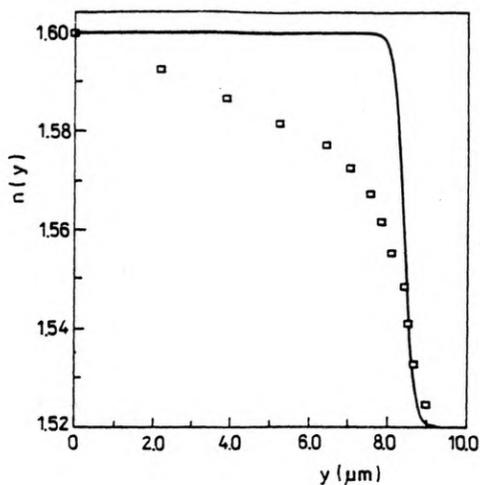
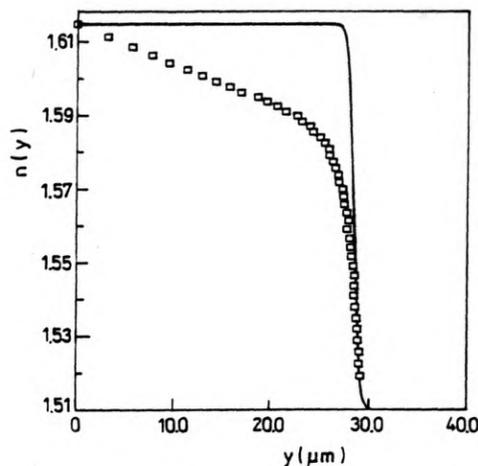
Fig. 9. Same as in Fig. 8, but for "D" type of glass ($T = 573 \text{ K}$, $t = 1 \text{ h}$)

phenomenon was observed in the case of "B" type glass. This glass contains in its composition less sodium (Tab. 1) which causes that the process of diffusion becomes slower, although the resulting change of the refractive index ($\Delta n = 0.08$) is comparable with that of the glass of "A" type.

The glass of "C" type, with the lowest percentage of sodium in its composition (Tab. 1), permitted the obtaining of waveguide structures with a small change of the refractive index ($\Delta n = 0.03$). However, in this glass, as well as in the glass of type "D", no dye penetration is observed. These glasses contain in their composition neither the Fe nor As compounds (Tab. 1). The result of this is a small material attenuation. In the case of the glass of "D" type it does not exceed 1 dB/cm . At the temperature $T = 573 \text{ K}$, this glass also has the highest coefficient of diffusion of Ag^+ ions. The change of the refractive index in this glass is the same as for the glass of the type "A". The results of the processes of diffusion carried out at the temperature $T = 573 \text{ K}$ for all the four types of glass are compiled in Table 2. It is clear therefrom that the coefficient of diffusion, as well as the obtained change of the refractive index depend on the contents of sodium in the given type of glass.

Table 2. Technological parameters of diffusion and electrodiffusion processes

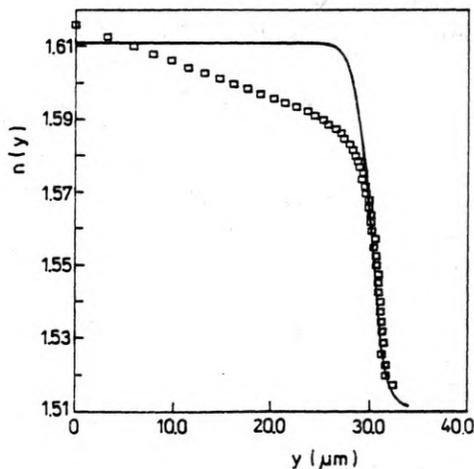
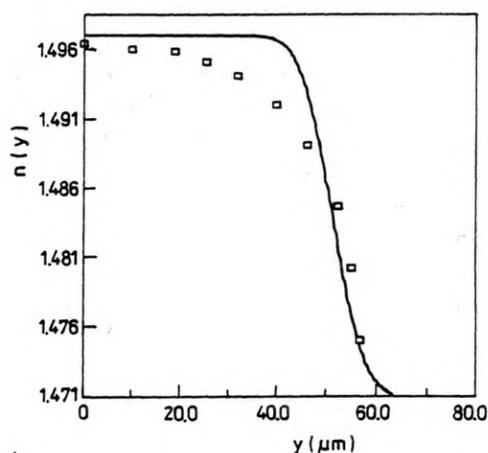
Glass	Na_2O [% mol]	D [$\mu\text{m}^2/\text{h}$]	Δn	r	f	n_s
A	12.21	6.8 ($T = 573 \text{ K}$)	0.1	0.1	1.5	1.511
B	8.44	0.7 ($T = 573 \text{ K}$)	0.08	0.2	1	1.520
C	3	9.6 ($T = 603 \text{ K}$)	0.03	1	4	1.471
D	15	12.76 ($T = 573 \text{ K}$)	0.1	0.5	5	1.511



▲

Fig. 10. Refractive index distribution in the glass of the type "A" after electrodiffusion of Ag^+ type ions ($T = 573 \text{ K}$, $E_0 = 36 \text{ V/mm}$, $t = 1 \text{ h}$). \square — experimental points determined by the IWKB method, solid line — numerical solution of Eq. (10)

Fig. 11. Same as in Fig. 10, but for the glass of the type "B" ($T = 623 \text{ K}$, $E_0 = 100 \text{ V/mm}$, $t = 0.5 \text{ h}$)



▲

Fig. 12. Refractive index distribution in the glass of the type "C" after electrodiffusion of Ag^+ type ions ($T = 603 \text{ K}$, $E_0 = 150 \text{ V/mm}$, $t = 2 \text{ h}$). \square — experimental points determined by the IWKB method, solid line — numerical solution of Eq. (10)

Fig. 13. Same as in Fig. 12, but for the glass of the type "D" ($T = 573 \text{ K}$, $E_0 = 40 \text{ V/mm}$, $t = 1 \text{ h}$)

The profiles of the refractive index obtained in the processes of electrodiffusion are exemplified in Figs. 10–13. When fitting the theoretical curves which are the solutions of Eq. (10) to the experimental points, the parameters D and r determined

in the processes of diffusion (Tab. 2) were assumed. Moreover, taking into consideration Nernst–Einstein compound

$$D_a = f \frac{kT}{e} \mu_a,$$

the coefficient of correlation f was estimated [3].

Worse fitting of the theoretical data to the experimental ones achieved for the processes of electrodiffusion may be explained by a different dependence of ion mobility on their concentration than that in the model assumed. The relatively best fitting was obtained for the glass of type "C" having the lowest sodium content. The calculated ratio of mobility for this glass is 1.

5. Conclusions

The comparative investigations of the exchange of Ag–Na ions, carried out for four types of glass, permitted the finding of a theoretical model describing the process of diffusion which is well fitted to the experimental data. In the case of electrodiffusion, the conformity of the experimental results with theory is worse, which fact may be explained by the different than assumed, dependence of ion mobility on their concentration. Moreover, a relationship between the coefficient of diffusion and the sodium contents in glass was found. Glasses of high sodium contents have also higher values of change of the refractive index with diffusion of Ag ions (Tab. 2). However, from the results obtained, it is not possible to conclude about the influence of sodium contents in glass on the value of the coefficients r and f . Low suppressibility of waveguide structures obtained in the glass of "D" type, and the high value of the coefficients D and Δn indicate a good usability of this type of glass as substrates for multimode structures.

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