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Detailed Study of the Two Steps for Fabricating $LiNbO_3$:Zn Optical Waveguides

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The two processes to fabricate planar and channel waveguides in congruent $LiNbO_3$ by vapor Zn diffusion are studied in detail. The first step, Zn-diffusion from its vapor phase, is controlled by the combination of temperature and time. The subsequent annealing step for the waveguide formation shows the typical dependence of the diffusion depth with time corresponding to a simple diffusion process. The diffusion coefficients show an Arrhenius dependence with the annealing temperature, with activation energies in the range 1.39–1.85 eV depending on the crystal cut.

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$LiNbO_3$ waveguides based devices have been intensively studied for several applications, including telecommunications systems,[1] and integrated[2] or nonlinear optics.[3] Different methods have been proposed for fabricating low loss optical waveguides in this material, for example proton exchange,[4] ion implantation[5] or metallic diffusion,[6] probably being the most popular technology the Ti-diffusion.[7] This technique preserves the high electro-optic, acustooptic, and piezoelectric coefficients of lithium niobate crystals. In addition, $LiNbO_3$:Ti waveguides present low propagation losses for both transverse electric (TE) and transverse magnetic (TM) measured in these channel waveguides (<0.1 dB/cm). However the titanium diffusion requires high temperatures (>1000 °C), and the waveguides suffer from high photorefractive damage,[8] which perturbs the propagating modes, degrading thus the device performance for high power operation. In order to avoid these disadvantages the use of Zn diffusion technology has been considered a good alternative.[9] This technique allows the fabrication of high quality optical waveguides (with losses in the order of 0.5 dB/cm) in both polarizations with low photorefractive damage.[9,10] Also, the temperature needed for Zn-diffusion is about 900 °C, lower than that the required for Ti-diffusion. There are two different approaches to fabricate $LiNbO_3$:Zn waveguides: the evaporation of a ZnO layer over a $LiNbO_3$ substrate[11] or the direct Zn incorporation on a $LiNbO_3$ substrate from its metallic vapor phase,[12] both followed by a thermal annealing. In recent years, using these methods, a wide variety of integrated optical devices have been developed, such as integrated lasers[13] or electrooptic modulators.[14] Nevertheless, up to date there is not as much information about the properties and the applications of zinc-diffused waveguides as for titanium diffusion or proton exchanged. In this work, a detailed study of the two-process fabrication of optical waveguides by Zn-diffusion from the vapor phase, in both *Z*- and *X*-cut, substrates is presented. For this purpose, the two steps involved in the process have been studied separately, making a technical study of the optimum parameters that can be controlled independently. Such parameters correspond to the diffusion time, diffusion temperature, external pressure, annealing temperature and annealing time.

The waveguide fabrication method consists of the Zn diffusion from the vapor phase, following a two step process as described elsewhere,[12] which has been performed in *Z*- and *X*-cut commercial wafers of lithium niobate crystals supplied by PHOTOX. With this procedure, low loss channel and planar waveguides which support both TE and TM propagation modes can be achieved.[12] Prior to the Zn diffusion, a lithographic process is done in one of the substrates faces,[15] based on the deposition of a SiO_2 layer, in order to define channel waveguides. The patterned oriented substrates are placed in a stainless steel tube reactor, cleaned in vacuum at high temperature (850 °C), where metallic zinc is deposited on the bottom. A connection in the upper part of the tube allows the system to be filled with Argon at a desired pressure. The tube is then introduced into an oven which fixes the necessary temperature during a controlled time, and finally a water circuit cools the top of the tube. So in this first step three different parameters can be controlled: temperature, pressure and time. This process implies the formation of a Zn-rich layer on the $LiNbO_3$ surface and through the channels opened in the SiO_2 mask. At the same time, the matrix suffers a chemical reduction which changes the colour of the samples to dark blue. To obtain useful optical waveguides a second step is necessary. This step consists of a thermal annealing of the sample performed in open atmosphere, by controlling the temperature and the time. The annealing step diffuses the Zn ions in the crystals, giving rise to channel waveguides in the patterned face of the sample and to a planar waveguide in the opposite face. In addition, the sample recovers the initial transparency due to a re-oxidation of the matrix. The waveguide fabrication finishes with the polishing of the two edges of the substrate, until optical quality is achieved for end-coupling purposes. The characterisation of the planar waveguides was performed by standard *m*-line spectroscopy, using a He–Ne laser (632.8 nm) coupled to the planar waveguide through an isosceles rutile prism pressed against it. With this technique, the refractive index distribution obtained after the two diffusion processes can be obtained. Also, light propagation in the channel and planar waveguides was examined by the end-fire-coupling technique with the aid of microscope objectives, using both TE and TM polarisation.

Several temperatures, between 500 and 800 °C, have been considered to perform the first step in the waveguide fabrication. While low temperature precludes the presence of enough vapor Zn and thus low Zn concentration in the superficial layer, high temperatures provoke excessive Zn incorporation and too thick layers. Therefore, an intermediate temperature of 550 °C has been chosen for the study, which allows obtaining a moderate film thickness with enough Zn concentration for the subsequent annealing step.

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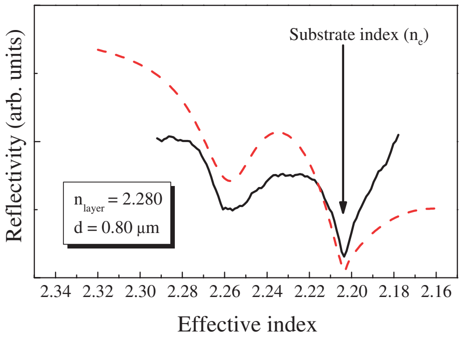


Fig. 1. Reflectivity curve obtained for a Zn-diffused *Z*-cut LiNbO$_3$ substrate using prism-coupling under TM polarised light (continuous line). Red dashed line corresponds to the theoretical fit using the parameters shown in the inset.

Once the diffusion temperature has been fixed, the effect of the pressure has been studied. Substrates diffused using different external controlled pressures (5.8, 58, and 580 mbar) showed very similar characteristics, indicating that the Zn concentration is controlled by the Zn vapor pressure, which is only temperature dependent. Therefore the external pressure was kept to 58 mbar in further studies. The influence of the diffusion time on the first step for waveguide preparation was also studied, and diffusion times in the range 0.5–2 h were examined.

The characterisation after this first step under *m*-line spectroscopy shows one or two broad modes for TM polarisation in *Z*-cut substrates and for TE-polarisation *Y*-propagating in *X*-cut substrates (Fig. 1). These facts are consistent with the formation of a high refractive index layer respect to the extraordinary index of the LiNbO$_3$ substrates, correlated with a layer of high Zn-content.[16)] When the light used for reflectivity measurements is TE-polarised in *Z*-cut substrates, or TM polarised in *X*-cut samples, no clear features are found, neither modes nor the critical angle corresponding to the refractive index of the substrates (ordinary refractive index). Using the reflectivity curve the optical parameters of the high index layer produced after the Zn-diffusion can be calculated by means of a computer program which models the reflectance of the multilayer structure.[12)] The modelling shows the formation of a step-index layer (Fig. 1), which acts as a waveguide when the polarisation corresponds to the extraordinary index of the substrate, allowing to obtain its refractive index value and thickness.

Figure 2 shows the layer refractive index as a function of the diffusion time using a temperature of 550 °C and a pressure buffer of 58 mbar, for the two different cuts (*Z*-cut: circles; *X*-cut: squares). As it can be observed, the refractive index of the Zn-rich layer for *Z*-cut substrates is nearly constant to a value of $n_{\text{layer}} \approx 2.26$, which is higher than the extraordinary index of the LiNbO$_3$ substrate (shown in the figure by a dashed line). By contrast, for *X*-cut samples, the refractive index of the layer is 2.26 for short diffusion times, and increases with the diffusion time. For $t = 4$ h, the refractive index of the layer is even higher than that of the LiNbO$_3$ ordinary index (also shown in the figure by a dashed line).

If the formation of the layer is a simple diffusion process, the depth layer should show a linear dependence with the square root of the diffusion time. Figure 3 shows the plot of the calculated layer depth as function of $t^{1/2}$. For *X*-cut samples this plot is linear in the whole range of time examined, while for *Z*-cut substrates the relation is linear up to 2 h of treatment, showing a sort of saturation for longer diffusion times. Considering the linear part of the plots, a diffusion coefficient can be obtained by using the expression:

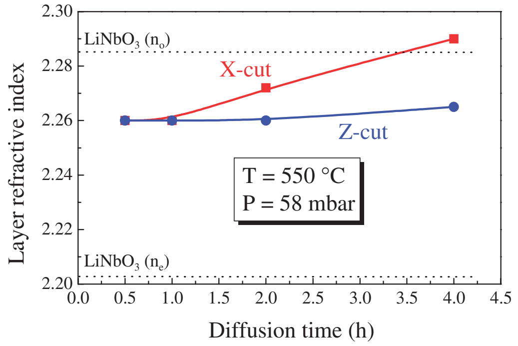


Fig. 2. Refractive index of the Zn-rich layer as function of the diffusion time for a temperature of *T* = 550 °C using a buffer pressure of 58 mbar (red squares: *X*-cut; blue circles: *Z*-cut).

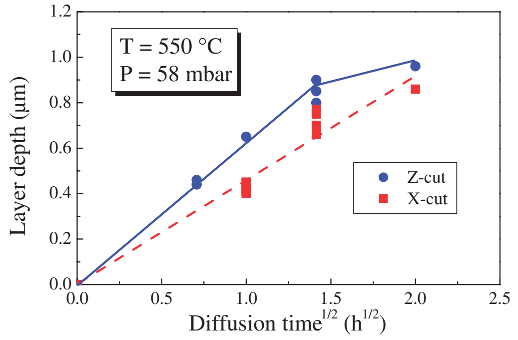


Fig. 3. Zn-rich layer thickness as function of the square of the diffusion time (red squares: *X*-cut; blue circles: *Z*-cut).

$$d = \sqrt{4 \cdot D \cdot t},$$

where *d* is the layer depth, *D* is the diffusion coefficient and *t* is the diffusion time. From the slopes of the plots in Fig. 3, diffusion coefficients of $D = 0.08\,\mu\text{m}^2/\text{h}$ for *Z*-cut, and $D = 0.05\,\mu\text{m}^2/\text{h}$ for *X*-cut substrates are obtained.

The second step in the waveguide fabrication corresponds to a thermal annealing of the Zn-diffused substrates. This study has been carried out at four different temperatures (800, 850, 900, and 950 °C), with annealing times ranging from 1 to 4 h, depending on the temperature. The time for each annealing temperature was chosen to obtain waveguides with similar parameters. To characterise the waveguides after this process, *m*-line spectroscopy and end-fire coupling technique were used.

By measuring the modal angles obtained by *m*-line technique, the index profiles were fitted to semi-Gaussian profiles on the form:

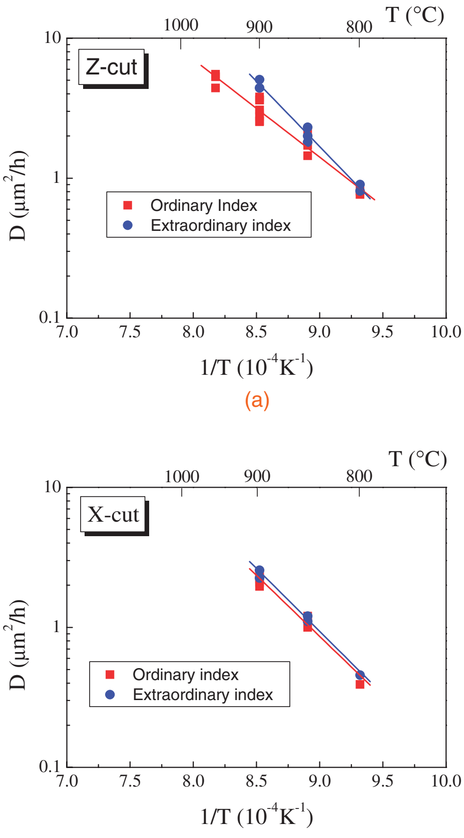
$$n = n_s + \Delta n \cdot e^{-x^2/d^2},$$

where n_s is the substrate refractive index, Δn the index increase and *d* the waveguide depth. The analysis of the index profiles after waveguide fabrication made for several treatment under Zn-vapor diffusion and the same annealing parameters (temperature and time) indicates that the waveguide parameters are very little dependent on the substrate treatment under Zn-vapor diffusion, as long as the Zn-layer is formed. Taking this fact into account, for studying the influence of the annealing step under different conditions for waveguide fabrication, the vapor-phase diffusion process parameters have been kept constant at $T = 550\,°\mathrm{C}$, $t = 2\,\mathrm{h}$ and $P = 58\,\mathrm{mbar}$.

Different annealing parameters (temperature and processing time) of two kind of oriented substrates (Z- and X-cuts) have been used to fabricate optical waveguides. It was not possible to excite waveguide modes by prism coupling for n_e polarisationf Z- and X-cut samples annealed at 900 °C during 2 h, due to the formation of a buried waveguide; in fact, light propagation using end-fire coupling was successfully achieved in these processed substrates. The buried waveguides can be due to the formation of a very thin low index layer at the surface of the sample, identified as ZnO compound by X-ray studies (refractive index 2.0 at 633 nm, lower than n_o and n_e of the lithium niobate crystal)[17] The refractive index changes (relative to the index substrate) of the graded index waveguides are in the range 0.14–0.30% for Δn_o and in the range 0.20–0.30 for Δn_e, with very little dependence on the crystal cut. These index increases, as well as the penetration depths, are well suitable to fabricate monomode optical waveguides in the visible and near infrared. It is worth to note that all these waveguides show optical propagation in both TE and TM polarisation by using end-fire coupling, either in the planar waveguides as well as in the different channel waveguides patterned in one face of the substrates. Although a systematic study of the losses has not been carried out in this work, typical reported losses in Zn-diffused waveguides are in the range 0.3–0.5 dB/cm for IR light.[11,18]

Figure 4 presents the dependence of the diffusion coefficients versus the inverse of the temperature, using the ordinary and extraordinary waveguides index profiles for Z-cut substrates [Fig. 4(a)] and X-cut substrates [Fig. 4(b)], where the data can be fitted quite well to a linear dependence. Taking into account this dependence, and assuming an Arrhenius law for the diffusion coefficients with the temperature, activation energies of $E_a = 1.39\,\mathrm{eV}$ for n_o and $E_a = 1.85\,\mathrm{eV}$ for n_e in Z-cut substrates were found. For X-cut substrates the activation energies were $E_a = 1.73$ and 1.78 eV for the n_o and n_e respectively. In the case of Z-cut substrates the strong difference between the two different activation energies could indicate two different mechanisms for the extraordinary index increase, such as some kind of Li-out diffusion. Finally, it is important to remark the higher diffusion coefficient found in Z-cut (0.7–6 $\mu\mathrm{m}^2$/h) respect to X-cut substrates (0.3–3 $\mu\mathrm{m}^2$/h) in the range of temperatures examined (800–950 °C).

The characterisation of optical waveguide fabrication by Zn diffusion from it vapor phase in congruent $\mathrm{LiNbO_3}$ substrates, by studying separately the two steps of the process, has been presented. The role of the different relevant parameters involved in the fabrication procedure (temperatures, times and external pressure) is established. The results show that the relevant parameter in the Zn-diffusion first-step concerns to the combination of temperature and time. The annealing second-step results show in most cases a typical dependence of the penetration depth on time, which corresponds to a simple diffusion process. The dependence of the diffusion coefficients as function of the annealing temperature indicates an Arrhenius law, with activation energies in the range 0.59–0.80 eV, depending on the crystal cut.



(a)

(b)

Fig. 4. Diffusion coefficients obtained from the refractive index profiles after the annealing step as function of the inverse of the temperature: (a) Z-cut substrates; (b) X-cut substrates.

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