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Proton-exchanged optical waveguides fabricated by glutaric acid

Wei Jun Liao*, Xianfeng Chen , Fucheng Chen, Yuping Chen, Yuxing Xia, Yingli Chen

Department of Physics, Institute of Optics & Photonics, Shanghai Jiao Tong University, 800 Dongchuan Rd., Shanghai 200240, China

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Abstract

In this paper, we first report that a new proton source, glutaric acid, has been used to fabricate optical waveguides in Z-cut lithium niobate crystals. The relationship was experimentally established between proton-exchanged (PE) waveguide parameters and fabrication conditions. It is shown that this new organic acid can be used to obtain deep PE waveguides in fast diffusion speed (0.275 $\mu m^2/h$ at 221°C) and with low loss (~ 0.2 dB/cm). It provides an alternative approach for fabricating PE waveguides in lithium niobate substrate.

Keywords: Proton exchange; Glutaric acid; Lithium niobate

1. Introduction

Lithium niobate, owing to its outstanding linear and nonlinear optical properties, has become a promising material to fabricate integrated optical components such as modulators, polarizers, filters and amplifiers [1,2]. The proton-exchange (PE) process, after firstly discovered in 1982 [3], has often been adopted to fabricate waveguides in lithium niobate wafers. It has been proved to be a simple and effective technique for forming high-index, low-loss lithium niobate waveguides. Proton exchange can be introduced to prepare waveguides in Z-cut, X-cut and Y-cut lithium niobate wafers [4,5]. In comparison to the commonly used Ti-diffusion technology [6], the PE process can obtain a high refractive index modulation with relatively low temperature and shorter exchange. The temperature varies from 150°C to 300°C and the time can vary between several minutes and dozens of hours. PE process can also get lower photorefractive damage and higher optical damage threshold for guiding a visible laser [7] than Ti-diffusion process.

Many weak acids have been adopted as proton sources to enhance the surface refractive index. Organic acids are most commonly used. Fabrications of planar waveguides and channel waveguides by using various organic acids have been reported, including benzoic acid [8–12], stearic acid [13], cinnamic acid [14], and pyrophosphoric acid [15]. Recently, the mixture of two organic acids [16] is also studied. Different phases of lithium niobate waveguides formed in these proton sources were investigated in details [17,18]. However, it is still very interesting to seek for more candidates to modulate the surface index of the wafer. In this paper, another organic acid, glutaric acid ($HOOC(CH_2)_3COOH$) is investigated for the first time, to our best of our knowledge. Glutaric acid has a melting point of 98°C and boiling point of 303°C, which is suitable for PE process. Additionally, it is involatile and safe to use. Optical waveguides formed by PE in the new proton source, as well as post-exchange annealing, are both experimentally analyzed. The results provide useful data to the practical applications of PE waveguides.

2. Waveguides by proton exchange

Twenty millimeters long, 10 mm wide and 1.5 mm thick Z-cut lithium niobate substrates with +Z surface polished were used for fabricating waveguides in our experiment. Before PE the wafers are bathed in boiling alkaline mixture ($1NH_4OH : 2H_2O_2 : 5H_2O$) for 10 min and then in boiling acid mixture ($1HCl: 2H_2O_2: 8H_2O$) for another 10 min in order to clean the surface. PE process is implemented in melting glutaric acid at 221°C with a temperature deviation less than 1°C, and the exchange time varies from 1 to 16 h. The ordinary refractive index will decrease after

* Corresponding author.
E-mail addresses: weijunliao@sjtu.edu.cn (W.J. Liao), xfchen@sjtu.edu.cn (Xianfeng Chen).

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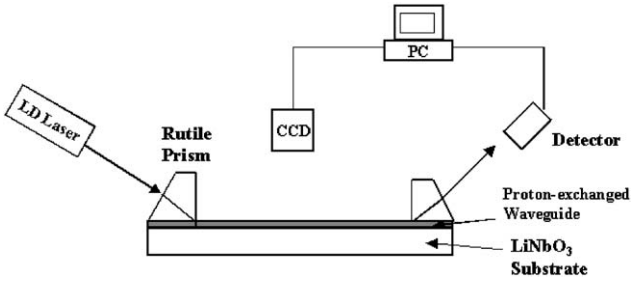

Fig. 1. Experimental setup for measuring the effective index and the propagation loss of PE waveguides fabricated in glutaric acid. Effective index of the waveguides are measured by bright m-lines method and the propagation loss is obtained from light propagation scattering method.

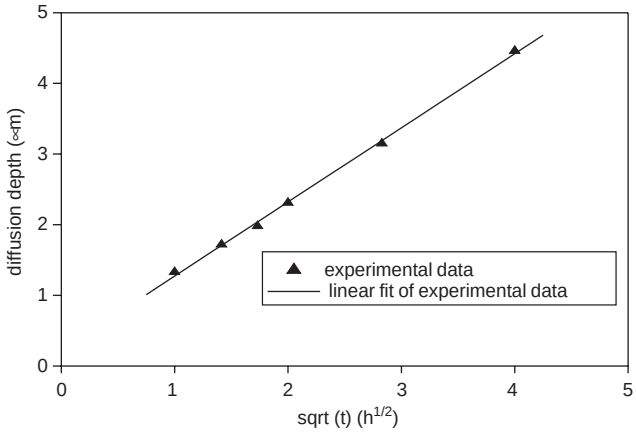

Fig. 2. Diffusion depth versus square root of exchange time at 221°C. It is shown that the depth d is proportional to square root of time $t^{1/2}$. Diffusion speed of glutaric acid at 221°C is obtained as $D(T) = 0.275$ μm^2/h by linearly fitting the experimental data.

proton exchange process, so that only TM guided-mode exists in Z-cut lithium niobate samples.

Bright m-lines [19] and light propagation scattering [20] methods are used to investigate the effective index and propagation loss of PE waveguides, as shown in Fig. 1. A diode laser with a wavelength of 650 nm is used. Two rutile prisms are adopted due to its higher refractive index than that of guided layer in substrate. The ordinary index n_o of rutile and the extraordinary index n_e of lithium niobate is 2.574 and 2.199, respectively at the wavelength of 650 nm. CCD camera is used to photograph the propagation scattering line of guided wave, and then the image is processed into computer to calculate the propagation loss of waveguide with a loss-measurement software. In comparison with other methods, the process in our loss-measurement has advantages of fast speed, no damage to waveguide and high precision as low as 0.1 dB/cm with deviation less than 0.05 dB/cm. We observed that the m lines from the PE waveguides by glutaric acid are similar as those from usually used benzoic acid. Up to 22th TM modes can be excited in the waveguide that was exchanged in glutaric acid for 16 h.

1, 2, 3, 4, 8 and 16 h are introduced as the exchange time to study the proton-exchange process of lithium niobate substrate in glutaric acid. It is shown by IWKB method that the formed PE waveguides also have a step index profile same as that exchanged in benzoic acid. Change of the extraordinary refractive index, waveguide depth and the loss calculated from the measurements with a 650 nm diode laser are as shown in Table 1.

From the table we can see that glutaric acid strongly modulate the surface index of lithium niobate substrate. It can be also found that the change of extraordinary index increases slightly with the exchange time owing to more abundant H^+ diffused into the sample. We believe the two abnormal points (0.113 at 4 h and 0.120 at 16 h) may come from the uncertainty of effective index measurement and the error of IWKB method. Many papers have proved that the relation between the diffusion depth and the exchange time can be expressed as

$$d = 2[D(T)t]^{1/2}, \quad (1)$$

where d is the diffusion depth, t is the exchange time, $D(T)$ is the diffusion coefficient at a given temperature. The equation shows that there is linearity between d and $t^{1/2}$, which can also be proved by our experimental data shown in Fig. 2. Diffusion speed of glutaric acid at 221°C is obtained as $D(T) = 0.275$ μm^2/h by linearly fitting the experimental data, which is rather fast. Extending the straight line to the

Table 1
The measured parameters of proton-exchanged waveguide in glutaric acid for 1, 2, 3, 4, 8 and 16 h

PE time t (h)	Depth of waveguide d (μm)	Change of extraordinary index	Waveguide loss for different mode (dB/cm)		
			TM_0	TM_1	TM_2
1	1.33	0.110	0.27	0.35	0.53
2	1.72	0.115	0.15	0.25	0.47
3	1.98	0.117	0.25	0.41	0.54
4	2.31	0.113	0.18	0.24	0.52
8	3.15	0.121	0.13	0.21	0.35
16	4.46	0.120	0.11	0.15	0.27

Table 2
Comparisons of PE waveguide parameters for benzoic acid [8], stearic acid [13], cinnamic acid [14], benzoic acid vapor [12] and glutaric acid (in this paper)

Proton source	Glutaric acid	Benzoic acid	Stearic acid	Cinnamic acid	Benzoic acid vapor
Δn	0.12	0.12	0.118	0.141	0.098
d (μm)	1.72 (2 h, 221°C)	1.3 (2 h, 220°C)	1.5 (2.5 h, 250°C)	1.62 (6 h, 220°C)	3.6 (24 h, 300°C)
$D(T)$(μm^2/h)	0.275(221°C)	0.207(220°C)	0.30(223°C)	Not mentioned	Not mentioned
Loss (dB/cm)	~ 0.2	2.4–4.8	1.5	0.8	0.35

Y-axis in Fig. 2, we found that it does not pass the origin point. We believe the reason is that the residual acid on the surface keeps exchanging with the substrate although majority of acid is separated from the wafer.

It is interesting to compare the PE waveguides fabricated in glutaric acid with those in other organic acids. The results are shown in Table 2. It can be found that glutaric acid is quite suitable for fabricating proton-exchange waveguides because the propagation loss is very small and the diffusion speed is fast.

3. Annealing process

PE waveguides with step-like index profile are always subjected to great loss when coupled to fibers, which will restrict the applications of PE waveguides in integrated optics. At the same time, annealing process will decrease the propagation loss further although the loss of PE waveguides by glutaric acid is rather low. It is also interesting and necessary to investigate the index redistribution character of PE waveguides fabricated in glutaric acid after annealing process. A number of papers have reported the annealing process using thin PE waveguides (0.15–1.1 μm). In our experiments, 3-h-exchanged PE thick lithium niobate waveguides fabricated in glutaric acid with a depth of 1.98 μm are annealed in air from 0.5 to 10 h at 350°C. After annealing the H^+ ion is redistributed, and the extraordinary index is gradually decreased as depth of waveguide increases. IWKB method is introduced to restore the index profile. The annealed proton exchanged (APE) waveguide by the new proton source has the similar graded-index profile in comparison with other reports [21,22], as shown in Fig. 3. From the curves in Fig. 3, we can see that the index distribution has been changed from step-like to more and more graduated profile as annealing time increases. Error function is introduced to fit the index profile of the APE waveguide as follows:

$$\Delta n_e(x) = \begin{cases} \Delta n_s, x \leqslant d_0, \\ \dfrac{\Delta n_s}{2\mathrm{erf}(d_e/d_a)} \left(\mathrm{erf}\left(\dfrac{d_e + (x - d_0)}{d_a} \right) \right. \\ \left. + \mathrm{erf}\left(\dfrac{d_e - (x - d_0)}{d_a} \right) \right), x > d_0, \end{cases} \quad (2)$$

where Δn_s is the change of surface index of APE waveguide, d_e is the depth of PE waveguide, d_a is the annealing depth,

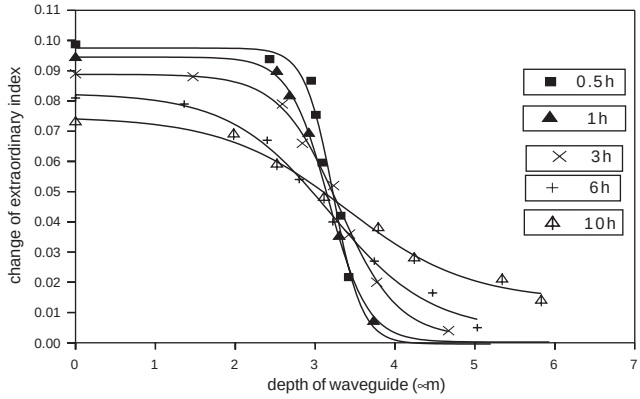

Fig. 3. Extraordinary index profile of guided-layer in APE waveguides with for various annealing hours. A 3-h-exchanged PE thick lithium niobate waveguides fabricated in glutaric acid with a depth of 1.98 μm are annealed in air from 0.5 to 10 h at 350°C.

Table 3
Properties of APE waveguides for different annealing time

Annealing time (h)	Δn_s	d_0 (μm)	d_a (μm)
0.5	0.098	1.3	0.4
1	0.094	1.2	0.6
3	0.089	1.1	0.9
6	0.081	1.0	2.0
10	0.073	0.7	3.5

d_0 is step depth decided by annealing time. Step depth d_0 results from that the PE samples are much thicker and will decrease as annealing time is increased. The parameters of APE waveguides annealed for different hours are shown in Table 3.

The surface index decreases when the annealing time increases. As shown in Table 3, it decreases by 0.04 when annealing time is 10 h. In the mean time, the annealing depth reaches to 2.0 μm with annealing time of 6 h. Further annealing will continue to lower the surface index and increase the depth of the waveguide. The propagation loss of the APE waveguide is less than 0.1 dB/cm limited by our loss measurement setup.

We have tried to exchange X-cut $LiNbO_3$ substrate with glutaric acid, but it is found that the surface of the wafer has been damaged, and no waveguide is found. The diffusion speed of proton exchange in X-cut $LiNbO_3$ substrate is faster than that in Z-cut sample [5], and the too fast

diffusion speed of glutaric acid in X-face lithium niobate substrate may attribute to the damage. We are now fabricating X-cut lithium niobate waveguide in glutaric acid mixed with lithium benzoate in order to reduce the diffusion speed.

4. Conclusion

Glutaric acid, a new proton source, has been used to fabricate PE and APE waveguides on Z-cut lithium niobate substrate. We found that the diffusion speed is fast in the new proton source, and lower propagation loss ($\sim$ 0.2 dB/cm) has been observed. Post-exchange annealing changed the index distribution of the waveguide from the step-like to more smooth profile. The experimental results show that glutaric acid may be a good proton source for fabricating PE and APE waveguides in integrated optics applications.

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