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Investigation on thermal properties of a new Nd-doped phosphate glass

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Abstract

Thermal properties of a new Nd-doped phosphate glass were investigated. The thermal expansion coefficient was measured by a dilatometric technique. A Mach–Zehnder interferometer was used to measure the temperature coefficient of refractive index (dn/dT) and the thermo-optic coefficient (ds/dT) in the temperature range of 30–100 °C. A low thermal expansion coefficient was observed for the new phosphate glass. The dispersion of the thermo-optic parameters was calculated from an empirical equation. The thermo-optic coefficient (ds/dT) at the working wavelength of 1053 nm is lower than those of other high average power glasses. These results indicate that the new Nd-doped phosphate glass is a good candidate for high average power laser applications.

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1. Introduction

Nd-doped phosphate glasses are used as host materials for high energy/high power (HEHP) laser applications because they posses high stimulated emission cross section and low nonlinear refractive index [1,2]. Another important application is for high average power (HAP) laser systems, which are predominantly used for laser peening of metals [3,4]. HAP laser glasses operate at thermal loading associate with high repetition rate. Thus, they require good thermal mechanical properties such as high fracture toughness, high thermal conductivity and low thermal expansion coefficient. These properties are commonly characterized in the well known thermal shock resistance which is expressed as [2]

$$R_s = \frac{K_{Ic}k(1-\mu)}{E\alpha} \tag{1}$$

where k is the thermal conductivity, μ is Poisson ration, K_{IC} is the

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fracture toughness, E is Young's modulus, α is the coefficient of thermal expansion.

However, phosphate glasses generally have a lower thermal conductivity, lower mechanical strength, but a higher thermal expansion coefficient than those of the silicate glasses and Nd: YAG [5]. The thermal conductivity of a glass is usually of the same order of magnitude, and normally lower than that of a crystal [6]. Thus, decreasing the thermal expansion coefficient is a possible way to improve the thermal mechanical properties of a laser glass.

Moreover, thermo-optic properties such as the temperature coefficient of refractive index (dn/dT) and the thermo-optic coefficient (ds/dT) are also important when considering the performance of laser systems. It is well known that ds/dT is an important thermo-optic parameter since it measures the distortions produced when the optical material is exposed to heat [7–9]. Such distortions could influence the laser cavity stability conditions, the output mode and reduce the laser beam quality. Therefore, a minimum ds/dT is desirable for optical applications as solid state lasers, where distortions of the laser rod should be avoided.

In this paper, we investigate the thermal properties of a new developed Nd-doped phosphate laser glass at Shanghai Institute of Optics and Fine Mechanics, named NAP2 Nd-doped

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phosphate glass. The refractive index, thermal expansion coefficients, dn/dT and ds/dT were measured. These glass parameters were discussed, and compared with other commercial Nd-glasses. Moreover, the dispersions of dn/dT and ds/dT were calculated.

2. Experimental

The chemical composition of NAP2 phosphate glass in wt% is as the following: P₂O₅ (55–65%), Al₂O₃ (6–18%), Li₂O (10– 18%), MgO (8–10%), BaO (0–5%), B₂O₃ (2–3%), La₂O₃ + Y₂O₃ + Nb₂O₅ + Sb₂O₃ (1–4%), SiO₂ (0–10%), Nd₂O₃ (0.5–4.0%)[10]. Glass samples with 0.5 and 3.7 wt% Nd₂O₃ were investigated in this paper, named NAP2-0.5 and NAP2-3.7, respectively. The used raw materials were first melted in a platinum crucible and stirred at 1350–1400 °C for a long time. After completing the melting process, the melt was quickly poured into a preheated steel mold and annealed for several hours at a temperature below T_g , the temperature of glass transition. To ensure the accuracy of parameters measured, all samples were annealed precisely for more than 10 days at a high temperature. The obtained samples were cut and polished into suitable shapes for different measurements.

The refractive indices of the glasses were obtained at 486 nm, 587 nm and 656 nm through V-prism method. And the refractive indices at 632.8 nm, 1064 nm and 1552 nm were determined by the Prism coupler method at room temperature, the uncertainty of these measurements is ± 0.0003 .

The thermal expansion coefficient was measured using a thermal dilatometer, Netzsch DIL402EP with a precision of 8 nm. The bulk sample with dimension of 5 mm \times 5 mm \times 20 mm was heated by 5 °C/min, up to the softening temperature point of the glass. The Raman spectra of the glasses were measured with a Renishaw InVia Raman spectrophotometer in 100–1500 cm⁻¹ spectrum range using the 488 nm excitation line.

The thermo-optic parameters were determined using a Mach– Zehnder interferometer with a 632.8 nm He–Ne laser. The samples are cylinders with 6 mm diameter and 50 mm length.

3. Results and discussion

3.1. Thermal expansion coefficient

Fig. 1 shows the thermal expansion length (dL/L_0) with increasing the temperature. We can calculate the linear thermal expansion coefficient by the expression below

$$\alpha = \frac{\mathrm{d}L/L_0}{\Delta T} \tag{2}$$

where ΔT is the temperature difference from room temperature to several other temperatures. From Fig. 1, it can be seen that the thermal expansion curves are essentially linear in the temperature range from room temperature to 480 °C, and no anomaly is observed when the measurement is carried out. At a certain temperature, the atoms in the glass start to move and the fluidity of the glass begins to increase. This corresponds to the glass transition temperature T_g . After this temperature the thermal expansion coefficient increases considerably. It is due

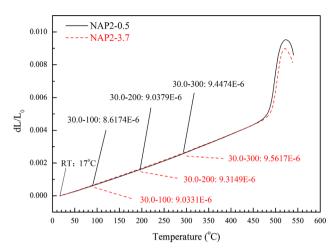


Fig. 1. Thermal expansion curves of NAP2 laser glass from room temperature (RT:17 $^\circ\text{C})$ to 550 $^\circ\text{C}.$

to that the atoms oscillate around their equilibrium position under thermal agitation. The thermal expansion coefficients from room temperature to 100, 200, and 300 $^{\circ}$ C are shown in this figure, too.

The thermal expansion coefficients of NAP2 laser glass and those of other Nd-doped phosphate glasses [11,12] are listed in Table 1. It can be observed that the thermal expansion coefficient of NAP2 laser glass is smaller than those of HEHP glasses, and is similar to those of HAP glasses [11].

It is well recognized that the thermal expansion coefficient of a glass depends on the connectivity of the glass network, and the thermal expansion is controlled by the asymmetry of amplitude of thermal vibrations in the glass. It decreases as the rigidity of glass network increases. An increase of the concentration of nonbridging oxygens (NBOs) would decrease the rigidity of structure and increase the coefficient of thermal expansion, whereas the changes of network former cation may cause either its increase or decrease depending on the effect on glass structure [13]. Therefore, the characteristics of the cations which form crosslink in the phosphate chains can determine the thermal expansion coefficient of a phosphate glass.

In [14], we have shown that one of the reasons for the smaller thermal expansion coefficient of NAP2 laser glass is the introduction of Li, which has larger field strength. Moreover, NAP2 laser glass also consists of MgO, B_2O_3 and SiO₂ components. Commonly, the replacement of BaO by MgO leads to a decrease of the thermal expansion coefficient [15,16]. The physical characteristic is explained by the corresponding values of the field strength (0.51 for Ba²⁺ and 0.95 for Mg²⁺ [15]). The larger field strength of the cations in the glass can lead to a tighter network.

On the other hand, the introduction of B_2O_3 into the phosphate glass network could decrease the thermal expansion coefficient [17–19]. Because it leads to a highly cross-linked glass structural network. In addition, the addition of SiO₂ to the phosphate glass can improve drastically the thermal expansion coefficient [20]. This is due to the silicate tetrahedra would increase the connectivity of the glass network. Because both P_2O_5 and SiO₂ are the network

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