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High refractive index $LaGaO_3$-TiO_2 amorphous spheres prepared by containerless solidification

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ABSTRACT

A series of amorphous spheres with nominal compositions of $(1-x)LaGaO_3$-$xTiO_2$ ($x = 0$, 0.625, 0.650, 0.675, 0.700, 0.725, 0.740) have been successfully prepared by containerless solidification. It is found that the $LaGaO_3$-TiO_2 system has an extremely wide glass-forming region of $0.625 \le x \le 0.740$; in this region, transparent amorphous spheres of diameter approximately 2–4 mm can be obtained. The maximum difference between T_g and T_x ($\Delta T = T_x - T_g$) exceeds 80 °C. The as-prepared amorphous materials exhibit a high refractive index of 2.49 for $x = 0.740$ and a transmittance exceeding 70% in the visible light range. The refractive index can be tuned by adjusting the amount of TiO_2. These findings show great promise for the application of $LaGaO_3$-TiO_2 systems as high refractive index materials and open up avenues for the exploration of novel potential optical components.

1. Introduction

Refractive index is an important physical parameter, and it provides a criterion for designing and developing novel optical components. Glasses with refractive indexes over 2.0 are useful for fabricating the lenses used in endoscopes, smart phones and the other potential applications. TiO_2 is well known as a colorless component with a refractive index higher than 2.55 [1], It is often used to enhance the refractive index of oxide glass. Although TiO_2 can be vitrified with network–forming oxides such as SiO_2, these network–forming oxides have very poor optical properties [2–6]. The refractive index of glass containing network–forming oxides is usually lower than the refractive indexes of La_2O_3–WO_3, BaO–TiO_2, and $LaO_{3/2}$–TiO_2 amorphous spheres prepared by containerless solidification without network-forming components [3,7,8].

Containerless solidification is an effective and efficient technique to vitrify oxides in bulk without adding any network-forming agent [9–15]. During the process of containerless solidification, bulk samples are levitated by gas without the container wall effect; this helps avoid inhomogeneous nucleation and leads to the development of deep undercooling melts [4].

In 2007, a $La_4Ti_9O_{24}$ amorphous sphere with a high refractive index of 2.231 at 1313 nm was prepared by containerless solidification without adding any network former [16]. In 2012, a series of TiO_2-based amorphous spheres were prepared successfully by containerless solidification without adding any network former; the highest refractive index realized was approximately 2.37 within the visible wavelength [17]. In 2015, TiO_2- and Nb_2O_5-doped La_2O_3 amorphous spheres with a refractive index of 2.29 in the visible light range were synthesized by containerless solidification [1]. Moreover, in 2017, La_2O-doped Ga_2O_3 amorphous spheres with a refractive index of approximately 1.955 for $(0.5La_2O_3$-$0.5Ga_2O_3)$ in the visible light range, which is consistent with the calculated value of 1.9 for $LaGaO_3$, were synthesized by containerless solidification [18,19].

To date, few studies focused on combining $LaGaO_3$ with TiO_2, which has a high refractive index of 2.55; such a combination is expected to yield a high refractive index. Therefore, in this study, a series of $(1-x)LaGaO_3$-$xTiO_2$ amorphous spheres, with nominal compositions for $x = 0$, 0.625, 0.650, 0.675, 0.700, 0.725, and 0.740, were prepared via containerless solidification. The region of formation and the optical, thermal properties of these spheres were investigated.

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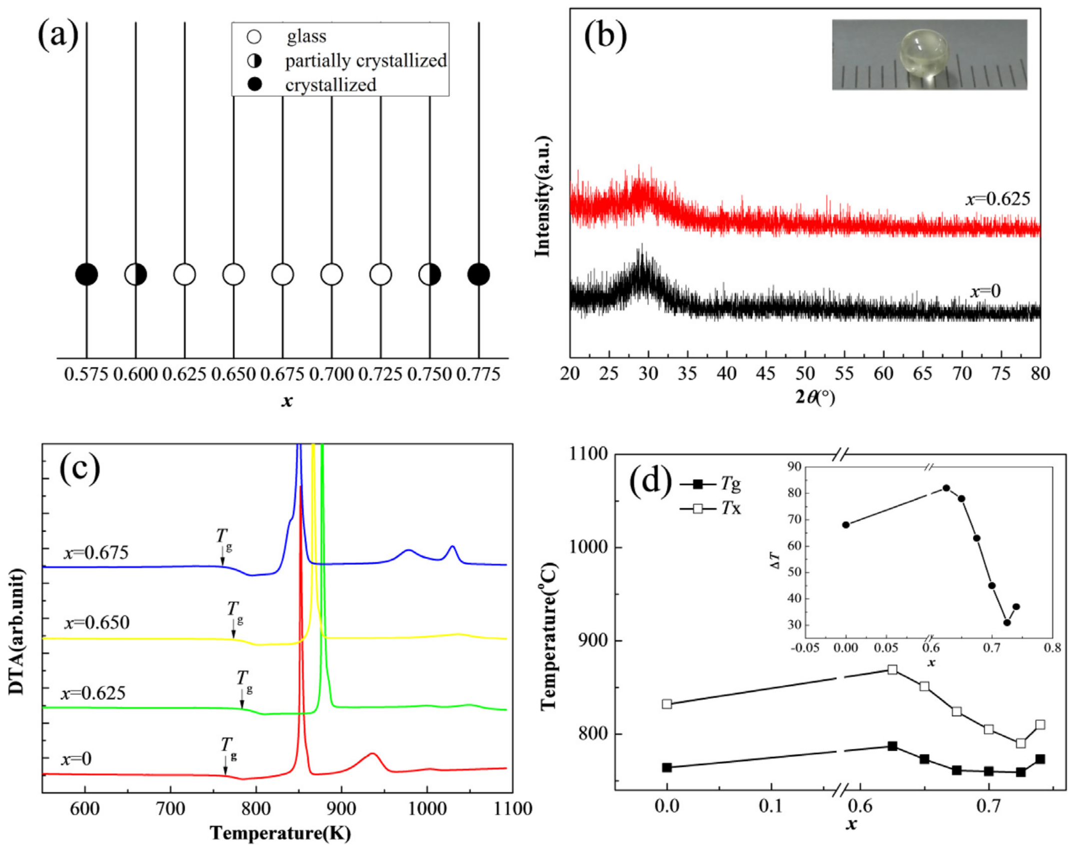

Fig. 1. (a) Forming region of the amorphous spheres. Open circles, half-filled circles and closed circles represent glass, partially crystallized, and crystallized, respectively; (b) XRD patterns of (1−x)$LaGaO_3$-$x$$TiO_2$ (x = 0, 0.625) amorphous spheres; (c) Differential thermal analysis curves of selected compositional samples; (d) X dependence of T_g, T_x and ΔT for the as-prepared amorphous samples.

2. Experimental

A series of amorphous spheres with the composition (1−x) $LaGaO_3$-$x$$TiO_2$ (x = 0, 0.625, 0.650, 0.675, 0.700, 0.725, 0.740) were successfully synthesized from TiO_2 powders (Sinopharm, China, >99.99%) and $LaGaO_3$ powders (prepared by sol–gel processes). The mixtures of powders were compacted into disks with a diameter of 10 mm and thickness of 1.5 mm prior to solid-state reaction in air at 1200 °C for 8 h. The sintered samples were melted in an aerodynamic levitation furnace, where transparent amorphous spheres could be obtained. Details of the aerodynamic levitation furnace are available referred in literature [4].

The structures of the amorphous spheres were observed by X-ray diffraction (XRD; SmartLab, Rigaku, Japan). The glass transition temperature (T_g) and the crystallization onset temperature (T_x), were investigated via differential thermal analysis at a heating rate of 10 °C/min (Setsys Evolution, SETARAM, France). Based on the Archimedes principle, the densities of the as-prepared amorphous spheres were measured by a device consisting of a four-digit electronic balance and its density component (AL104, Mettler Toledo, China). The amorphous spheres were optically polished into discs for optical tests carefully. The transmittance spectra of the amorphous spheres were observed using a UV–vis spectrometer (Cary 5000, Varian, USA). The characteristics of the refractive indexes of the samples were evaluated by using an SPEC ellipsometer (SE 850 DUV type, SENTECH, Germany).

Table 1
x dependence of densities for as-prepared amorphous spheres (the density of the distilled water is 0.9982 g/cm^3 at 20 °C).

x	Dry weight	Wet weight	Δ weight	ρ (g/cm^3)
0	0.3337	0.2777	0.0560	5.948
0.625	0.3300	0.2668	0.0632	5.212
0.650	0.4967	0.4008	0.0959	5.170
0.675	0.3785	0.3049	0.0736	5.133
0.700	0.4450	0.3568	0.0882	5.036
0.725	0.3607	0.2882	0.0725	4.966
0.740	0.2654	0.2117	0.0537	4.933

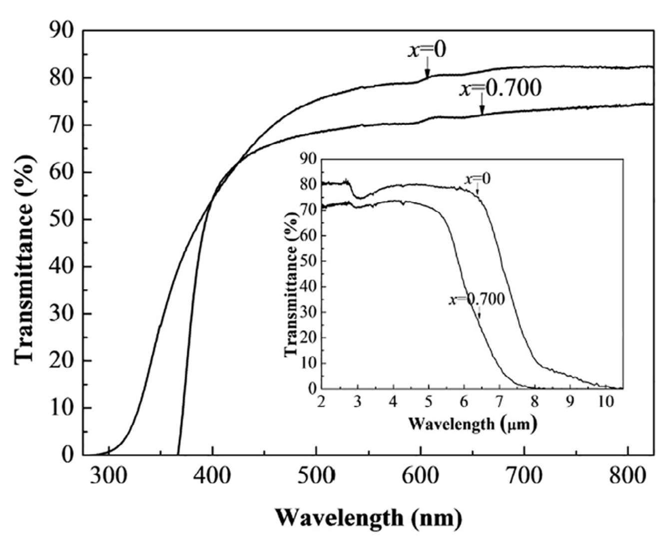

Fig. 2. Transmittance spectra of the typical compositions of as-prepared amorphous spheres with x = 0 and 0.700 in the UV–vis and IR region.