

The Effect of Sintering Temperature and Soaking Time on the Transparency of Silica Glass Prepared by Gel-Casting and Sintering

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Abstract

Transparent silica glass was synthesized by a novel green and simple route involving gel casting of fumed silica nanoparticles followed by solid-state sintering. The effect of sintering temperature and soaking time on the transparency of the samples were investigated. Three temperatures of 1000, 1100 and 1200°C and five soaking time of 25, 45, 60, 120, 180 min were examined. The prepared samples were characterized with different techniques including XRD, FTIR spectrometry, and UV-VIS spectrophotometry. The results showed that highly transparent silica glass, with transparency of 83%, can be obtained by sintering at 1100°C for 180 min with heating rate of 5 °C/min. The absence of the isolated silanol and hydroxyl group, the amorphous structure, and the low porosity are the reasons behind the high transparency. The glass has high density of 2.32 g/cc and high microhardness of 7.30 GPa.

Keywords: Transparent Silica Glass, Gel-Casting, Fumed Silica, Agar, Solid-State Sintering

INTRODUCTION

Silica glass attracts fundamental interest as a model for glassy material because of its relative structural simplicity and high purity. Further, it is an outstanding optical material due to its high transparency in the near infrared to vacuum-ultraviolet regions of the light spectrum [1].

Glass materials are often produced by traditional melting method; however, this method isn't suitable for the silica glass as it consumes high energy due to the high melting point of pure silica. Thus, many alternative methods have been reported to synthesis silica glass at low temperatures and/or short time. These includes the vapor-phase axial deposition method [2,3], the sol gel method followed by sintering [4-9] and spark plasma sintering [10-12].

Although these methods overcome the main drawback of the melting method, each of them, however, has its own limitations regarding productivity, cost, ease of application, transparency, purity, the maximum producible thickness, and the ability to produce complex shapes. The flame fusion method, for example, produces silica glass with relatively

high impurities and requires high skilled craftsmen [13]. On the other hand, the sol gel method, in addition to its high cost and complexity, results in silica glass bodies usually contain a large amount of OH groups (>1000 ppm), thus, further heat treatment under vacuum or in an atmosphere containing He and/or Cl₂ is required [4,14]. The spark plasma sintering and vapor-phase axial deposition are costly, complex, and unsuitable for producing complex shapes [3,10,11].

Recently, the solid-state sintering has been reported to be suitable for synthesis transparent silica glass from PVA/SiO₂ nanocomposite at relatively low temperatures, however, the followed procedure is complex, time consumable, and restricted to thin and flat shapes [15,16].

In our previous work [17], a novel green and simple route, involving gel casting followed by solid-state sintering, was developed to synthesis transparent silica glass. It has been found that this route has the potential to overcome the drawbacks directed against the other techniques. It combines the advantage of the melting method, i.e. molding into complex shapes, with the advantages of the solid-state sintering. In the current work, the effect of sintering temperature and the soaking time on the transparency of the synthesized transparent silica glass was investigated.

MATERIALS AND MOTHEDES

Transparent silica glass was prepared from a suspension of fumed silica nanoparticles (surface area 380 g/cm³, Guangzhou GBS High-Tech.& Industry Co. Ltd, Guangzhou, China) in aqueous solution of agar (HiMedia Laboratories Pvt. Ltd.) as described in details elsewhere [17]. Briefly, the suspension was prepared by adding 3g of fumed silica nanoparticles to 78 ml of distilled water containing 0.15g of agar. It was mixed by magnetic stirrer at room temperature and treated with sonication for 3h. The agar in the suspension was dissolved by heating the suspension in a microwave for 150 sec with a power of 900W and a frequency of 2450 MHz. The resulting viscous slurry was mixed on a magnetic stirrer for around 10 min till its temperature reaches 55°C when it was casted into PVC mold placed on a substrate of plaster. After few hours of drying at room temperature in air, the samples were demolded and dried in oven at 55°C. The green

bodies were heat-treated at 600°C for 2h at a rate of 5°C/min to remove the agar. The sintering was achieved at 1000, 1100, and 1200°C for 25, 45, 60, 120, and 180 min with a heating rate of 5°C/min. The sintered samples were gradually grinded with SiC paper (up to grit 2000) and polished with SiO₂ slurry before the characterization.

XRD pattern of the samples was recorded using X-ray diffractometer (XRD 6000, Shimadzu, Japan). The microstructure of the green body and the sintered sample was examined using SEM (Inspect S50/ Japan). Infrared spectra of fumed silica powder and heat-treated sample were acquired using a Fourier transform infrared (FTIR) spectrometer (Shimadzu 1800, Japan) over a wavenumber range of 4000–400 cm⁻¹. Transmittance of the sintered samples was measured using (SHIMADZU, UV-1800, Japan). The microhardness of the dense samples was tested using Digital Micro Vickers Hardness Tester (TH-717). The density of the sintered samples was measured using the Archimedes method.

RESULTS AND DISCUSSION

Fig (1) shows the result patterns of the x-ray diffraction analysis of the samples sintered at different temperatures. The broad peak for the samples sintered at 1000°C and 1100°C indicates the amorphous nature, i.e. the short range order, of the samples [13,18]. The XRD pattern of the sample sintered at 1200°C shows sharp peaks assigned to (101), (111), (102), (200), (211) and (301) planes of tetragonal cristobalite in full agreement with JCPDS No. (00-011-0695).

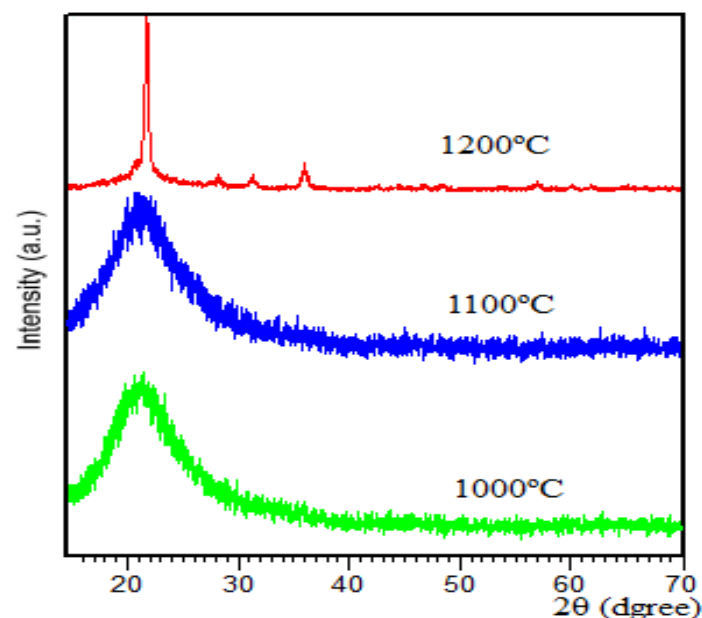


Figure 1: XRD patterns of sample sintering at (1000, 1100, 1200°C) for 3h with rate 5°C/min

Fig (2) shows the photograph images of the samples prepared

via gel casting method followed by sintering at different temperatures. It is obvious that the samples sintered at 1000°C aren't transparent indicating that the process of sintering is incomplete and the leak in the transparency is due to the high porosity [10]. As for the samples sintered at 1200°C, the opaqueness is due to the formation of the cristobalite crystalline phase as confirmed in Fig (1). The samples sintered at 1100°C exhibit an interesting transparency indicating the high densification, low porosity, and leak of crystallinity.

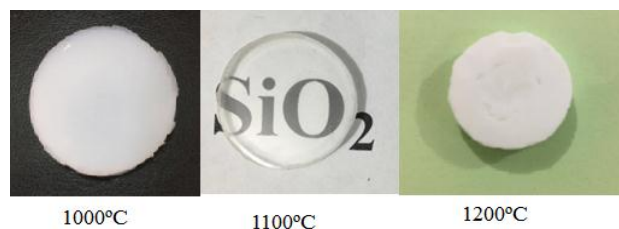


Figure 2: Photograph images of the samples sintering at different temperatures

Fig (3) shows the FT-IR spectra of the sample sintered at 1100°C. The peak near 466 cm⁻¹ characterizes the bending vibration of Si-O-Si bonds [14,15,19]. The band about 796 cm⁻¹ is related to the Si-O-Si symmetric stretching vibration mode of bridging oxygen between tetrahedral [14,15,19]. The peak around 1097 cm⁻¹ is corresponding to Si-O-Si asymmetric stretching of bridging oxygen between tetrahedral of bridging oxygen within the tetrahedral [14,15,19]. It is important to note that the peak of the silanol group (Si-OH) at 976 cm⁻¹ isn't observed for the sample sintered at 1100 °C. Similarly, the hydroxyl group for H₂O at 3433 and 1636 cm⁻¹ were also not observed. These groups have the ability to inhibit the transparency of the silica glass. [15]

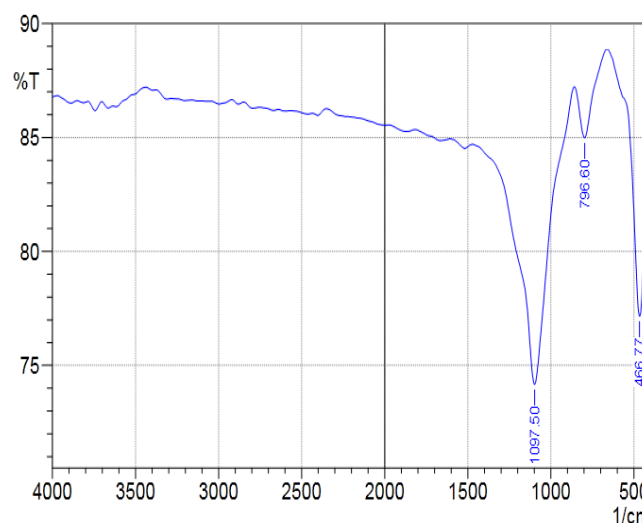


Figure 3: FTIR spectra of the transparent silica glass sintered at 1100°C for 3h

Fig (4) shows the UV-VIS spectra of the samples prepared by sintering at 1100°C for different soaking times. It can be observed that the transmittance increases with the increase of soaking time. The raise of transmittance is due to the more densification of the sample as evidenced by the increase of density and the decrease of porosity as shown in Fig (5) and (6) respectively.

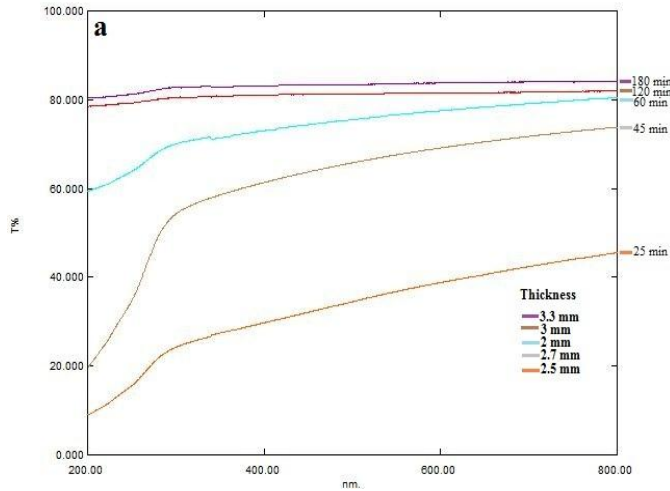


Figure 4: UV-VIS spectra of samples sintered at 1100°C for different soaking times

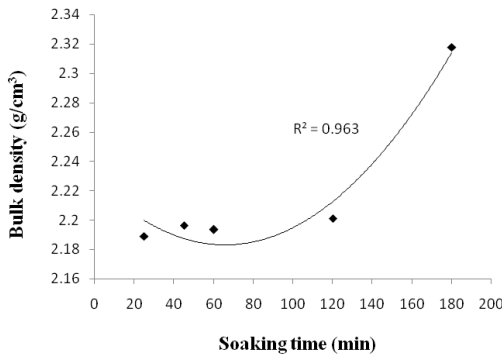


Figure 5: Bulk density of sample sintered at 1100°C for different soaking times

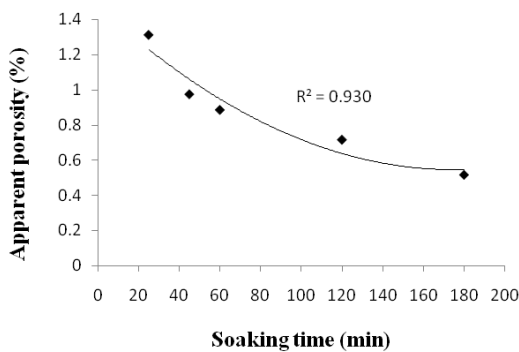


Figure 6: Apparent porosity of sample sintered at 1100°C for different soaking times

The highest transmittance value was obtained for the sample sintered at 1100°C for 180 min; its transmittance was 82.9% at wavelength 350 nm with thickness 3.35 mm. This sample has the highest bulk density of 2.32 g/cm³ and the lowest porosity of 0.5%. The density increases with the increase in soaking time due to the convergence of particles, which allows full coalescence of the particles and induces the viscous flow which are necessary for the densification [10]

Fig (7) shows microhardness values for the samples sintered at 1100 at different soaking times. The highest hardness of 7.30 GPa was obtained for the sample sintered for 180min, this is because when soaking time increases the defects, which cause deteriorates in mechanical properties, are reduced.

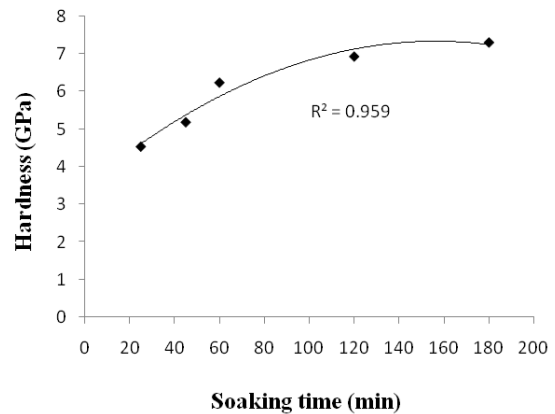


Figure 7: Microhardness of sample sintered at 1100°C for different soaking times

CONCLUSIONS

Green bodies of fumed silica glass made by gel casting method were sintered at different temperatures and soaking times. The highest transmittance was obtained for the sample sintered at 1100°C for 180 min. The high transparency results from the amorphous structure, absence of silanol and hydroxyl groups, and the low porosity.

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