Realizing translucency in aluminosilicate glass at ultralow temperature via cold sintering process

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Abstract: Glass with high visible-light transparency is widely considered as the most important optical material, which typically requires a processing temperature higher than 1000 °C. Here, we report a translucent aluminosilicate glass that can be prepared by cold sintering process (CSP) at merely 300 °C. After eliminating structural pores in hexagonal faujasite (EMT)-type zeolite by heat treatment, the obtained highly active nanoparticles are consolidated to have nearly full density by adding NaOH solution as liquid aids. However, direct densification of EMT powder cannot remove the structural pores of zeolite completely, leading to an opaque compact after the CSP. It is proved that the chemical reaction between the NaOH- and zeolite-derived powders is highly beneficial to dissolution–precipitation process during sintering, leading to the ultra-low activation energy of 27.13 kJ/mol. Although the addition of 5 M NaOH solution greatly promotes the densification via the reaction with aluminosilicate powder, lower or higher concentration of solvent can deteriorate the transmittance of glass. Additionally, the CSP-prepared glass exhibits a Vickers hardness of 4.3 GPa, reaching 60% of the reported value for spark plasma sintering (SPS)-prepared sample.

Keywords: glass; cold sintering process (CSP); optical properties; mechanical properties

1 Introduction

Among various attractive properties of glass materials, the optical function is widely considered as the most important one [1], which mainly relies on the transparency of the glass in the visible-light spectrum [2,3]. However, the conventional technology of preparing the glass materials typically requires a temperature as high as 1200 °C, which is highly energy-consuming in modern industry [4]. For saving energy, field-assisted sintering has been proposed as an advanced technology for preparing the glass materials, due to the relatively low processing temperature compared to conventional methods. In particular, spark plasma sintering (SPS) technique can not only decrease the densification temperature [5] but also improve the transmittance compared with pressure-less sintering [6]. Moreover, it has been found that, by using mesoporous powders with high sintering activity, the densification temperature of transparent glass materials can be reduced to lower than 1000 °C.
Nomenclature

| Acronym | Description |
|---------|-------------|
| EMT     | Synthesized EMT powder (uncalcined) |
| C-EMT   | 550 °C calcined EMT powder |
| ZC      | Cold-sintered EMT with NaOH solution (zeolite ceramic) |
| DP-glass| Dry-pressed C-EMT under certain temperature |
| DW-glass| Cold-sintered C-EMT with deionized water |
| NS-glass| Cold-sintered C-EMT with NaOH solution |

[7,8]. Recently, a novel sintering technique named cold sintering process (CSP) has received intensive attention owing to the ultralow temperature (typically around 300 °C) for densification [9–16]. During the CSP, when suitable liquid aids are selected to dissolve the edges of the particles, the pores between the particles can be eliminated by the evaporation of the solvent and precipitation of the solute [17,18]. So far, many oxide materials including ZrO2 [19], MgO [20], Bi2O3 [21], CaCO3 [22], and SiO2 [23–27], have been successfully prepared by the CSP. However, very few studies related to the CSP-densified glass materials have been demonstrated [28]. More importantly, there has been no cold-sintered glass that can display certain transmittance in the visible-light spectrum for possible optical applications.

In this study, we report the translucent glass prepared by the CSP using a highly active raw powder derived from hexagonal faujasite (EMT)-type zeolite for the first time. The EMT-type zeolite is a hexagonal polymorph of the cubic FAU-type (faujasite structure) zeolite that plays a very important role in catalysis [29]. Due to the high Al/Si ratio and surface area, the EMT powder shows weak thermal stability upon heating. Although the CSP of several zeolite powders, such as 4A, 13X, and Y types, has been demonstrated before [30], dense glass materials with certain transmittance have never been achieved, highlighting the importance of this study.

2 Materials and methods

2.1 Material preparation

The EMT zeolite was synthesized by following the reported procedure [31]. The as-synthesized EMT powder of 2.5 g was added to NH4Cl solution and stirred for 1.5 h for ion exchange. After repeated washing by centrifuge and freeze-drying, the ion-exchanged EMT was calcined in a muffle furnace at 550 °C for 3 h, which is denoted as C-EMT. For the CSP, NaOH solution (5 M) of 240 µL was added to the C-EMT powder of 0.6 g, and then ground to form a uniform mixture. The mixed powder was transferred to a hand press with a circular heated mold (Shanghai Jingsheng Scientific Instrument Co., Ltd., China), which was manually applied with a pressure ranging from 300 to 500 MPa. The mold was made from hot work steel, and the inner diameter of the die was 10 mm. Various temperatures of 100–350 °C and holding time of 10–50 min were used for the CSP. Based on different liquid aids, the sintered samples are denoted as NS-glass (NaOH solution), DW-glass (deionized water), and DP-glass (dry press). For comparison, the EMT powder was cold sintered with the NaOH solution directly without calcination, and the sample was denoted as ZC.

2.2 Characterization

The densities of the bulk samples were measured by Archimedes’ method. X-ray diffraction (XRD) patterns of the samples were measured with an X-ray diffractometer (Dandong HAOYUAN Instrument Co., Ltd., DX-2700B, China; 40 kV, 200 mA, Cu Kα1, k = 1.54056 Å). The microstructures of the samples were observed by a field emission scanning electron microscope (FE-SEM; TESCAN, MAIA3, Czech Republic). Transmission electron microscopy (TEM) measurements were conducted on a transmission electron microscope (JEOL, JEM-2100F, Japan). The valence states of the elements in power and bulk materials were recorded by an X-ray photoelectron spectrometer (Thermo Scientific, Escalab 250Xi, USA). The binding energy was calibrated based on the line position of C 1s (284.8 eV). Fourier transform infrared (FTIR) spectra were collected by an infrared imaging spectrometer (Nicolet, In10MX, USA). Thermogravimetric (TG) analysis and differential scanning calorimetry (DSC) tests were performed by a thermal analyser (NETZSCH, STA449, Germany) from room temperature to 800 °C in the air with the heating rate of 5 °C/min. The in-line transmittance (%) was measured by an ultraviolet–visible–near-infrared spectrometer (Shimadzu, UV3600, Japan); the detection wavelength range is 300–900 nm, and the sample’s thickness is 1 mm.

For the mechanical testing, the double-side polished ceramic discs with a diameter of 10 mm and a thickness of 1 mm were used. Young’s modulus and Poisson’s ratio were determined by an advanced ultrasonic material characterization system (TACLAB, UMS-100), the voltages for testing shear and longitudinal wave
velocities were set as 200 and 50 V, respectively, the probe frequency was 20 MHz, and the gain of the curve was set as 15 dB; every single sample was tested for 4 times. The Vickers hardness was obtained by the standard indentation method under a load of 9.8 N for 5 s. The flexural strengths (\( \delta \)) of the samples were measured by the modified small punch (MSP) test [32]:

\[
\delta = \frac{3P_{\text{max}}}{2\pi^2 t} \left[ 1 - \frac{1}{4} \frac{b^2}{a^2} + \ln \frac{a}{b} \right]
\]

where \( P_{\text{max}} \) is the failure load, i.e., the maximum load to destroy the sample; \( \nu \) is the Poisson’s ratio; \( a \) and \( b \) are the radius of the hole in the bearing die and the radius of the cylindrical indenter, respectively; and \( t \) is the thickness of the sample.

### 3 Results and discussion

As shown in Fig. 1(a), the as-synthesized EMT powder has an ultrafine plate-like feature with an average particle size of lower than 50 nm. High-resolution TEM (HRTEM) image reveals that the nanocrystals mainly exist in the form of hexagonal molecular sieves, which have highly ordered channels (the inset of Fig. 1(a)). The XRD pattern of this powder with broadened Bragg peaks can be indexed to the hexagonal EMT structure in the \( P63/mmc \) space group (Fig. 2), which is in agreement with the TEM observation. Although the low framework density of the EMT zeolite leads to low thermal stability, the temperature of the CSP is not high enough to completely collapse such structural pores, which could deteriorate the transmittance significantly due to the scattering effect on light. Therefore, the heat treatment is necessary to eliminate those structural pores while preserving the ultrafine grain size of particles. According to the TG measurement, there was a weight loss of as much as 27.5% until 550 °C, but no noticeable changes occurred with further increasing temperatures. The weight loss can be mainly ascribed to the release of NH\(_3\) [33], though the exothermic peak (Fig. 1(b)) at 350 °C should be attributed to the collapse of the ordered porous structure in the EMT powder, which was confirmed by the XRD (Fig. 2, 350 °C-EMT). The TEM image shows that there is no ordered porous structure observed in the C-EMT, while the particle size of the C-EMT only increased mildly compared with that of the pristine EMT (Fig. 1(c)). The FTIR spectra (Fig. 1(d)) further indicate that the as-synthesized EMT has peaks at 664 and 743 cm\(^{-1}\), corresponding to the internal tetrahedral symmetric stretching and external link symmetric stretching vibrations [34], respectively. The appearance of the peak at 729 cm\(^{-1}\) indicates the formation of newly tetrahedral aluminum atoms with the collapse of the framework [35]. In addition, the peak at 857 cm\(^{-1}\) can be assigned to SiO\(^{−}\) and AlO\(^{−}\) defects in the framework structure [36]. The disappearance of these peaks strongly suggests the collapse of the EMT framework after calcination, which is consistent with the TEM and XRD results (Fig. 2(a)). However, the peak at 979 nm\(^{-1}\) is considered to be the asymmetric vibration caused by internal Si–O–Al (T–O–T), which can still be found in the C-EMT despite a shift probably due to the change of coordination environment.

The thermal conversion of EMT to C-EMT has a significant influence on the resultant structure of compacts after the CSP, which can be revealed by the XRD results clearly (Fig. 2). When the EMT was directly utilized as the starting powder, all the peaks belonging to the EMT changed to a cubic structured zeolite (Na\(_6\)(AlSiO\(_{4}\))\(_{6}8\)H\(_2\)O, PDF Card No. 40-0102) after the CSP at 300 °C under 500 MPa by using the NaOH solution as the liquid aids. This result is very different from Ref. [30] for cold-sintered 4A and 13X zeolites, which showed the completely same zeolite structure after the CSP. On the one hand, the CSP-induced phase transformation reflects the weak stability of the EMT compared to other zeolites. On the other hand, it confirms that the CSP alone could not eliminate all the structural pores in zeolite. As a result, the final density of cold-sintered zeolite ceramic (denoted as ZC) was 2.25 g/cm\(^3\). In comparison, the CSP of amorphous C-EMT powder by using the same conditions gave rise to a glass pellet (denoted as NS-glass) with a density of 2.41 g/cm\(^3\). If we assume the density of the aluminosilicate glass prepared by the SPS at 780 °C as that of the fully dense sample (2.52 g/cm\(^3\)) [31], the relative density of the NS-glass has reached 95.6%.

The distinct densification effect by using different powders and liquid aids can be directly observed from the fracture surfaces of the bulk samples sintered at 300 °C under 500 MPa for 30 min (Fig. 3 and Fig. S1 in the Electronic Supplementary Material (ESM)). The fracture surface of the ZC sample (Fig. 3(d)) shows a generally dense but rough surface with noticeable pull-out grains in contrast to the porous fracture surface of the DP-glass (Fig. 3(a) and Fig. S1 in the ESM).
Fig. 1 TEM images of (a) EMT and (c) C-EMT powders. Inset of (a) is the HRTEM image of EMT zeolite particle. (b) TG–DSC curves of EMT powder. (d) FTIR spectra for EMT and C-EMT powders.

In comparison, a much more homogeneous fracture surface can be observed by using the C-EMT powder for the CSP, which is a typical feature of the glass material. When water was added as the liquid aids (denoted as DW-glass), many individual particles on the fracture surface could be distinguished despite the absence of too large pores, and the grain size increased to 30.75 nm (Figs. 3(b) and 3(e)). The phenomenon of the grain growth can also be observed in the NS-glass (Figs. 3(c) and 3(f)), where the fracture surface shows almost pore-less feature with a larger grain size (35.49 nm) compared to that of the DW-glass, suggesting that an intermediate dissolution–precipitation procedure took place via the reaction. Since the zeolites [37,38] are frequently synthesized by hydrothermal methods, during which the NaOH solution was introduced to react with the aluminum and silicon compounds through the joint action of temperature and pressure in a closed reactor, and the environment for preparing the NS-glass via the CSP with pressure and solution is similar to the hydrothermal conditions. Thus, it is reasonable to deduce that the densification of amorphous aluminosilicate can be accelerated by the analogous process for synthesizing zeolite powder [30,37,39]. Surprisingly, there is no obvious Na-rich position on the fracture surface of the NS-glass (Figs. 3(g)–3(i)). Although the total atom ratio of Na element in the NS-glass is higher than that of the DP-glass (Table S1 in the ESM), the distributions of the Na element in both samples are almost the same (Fig. S2 in the ESM), suggesting a homogeneous reaction of NaOH with the C-EMT powder.
To understand the densification process of the C-EMT powder, the influence on concentration, temperature, pressure, and holding time with different liquid aids were investigated. First of all, it can be seen that the liquid aids played a critical role in the CSP. The samples sintered with the NaOH solution (5 M) shows the highest density in any conditions (Figs. 4(a)–4(d) and Fig. S3 in the ESM). In addition, both NaOH solution and water can promote the densification of C-EMT compared to the samples sintered with bare powder, especially for the increased concentration of the NaOH solution. Therefore, it is deduced that the C-EMT powder can react with both NaOH and H₂O, noting that the pristine EMT mentioned above is normally composed of Na₂O–SiO₂–Al₂O₃–H₂O. Moreover, higher temperature and pressure can further facilitate the reaction between powder and solution, leading to the highest density of around 2.41 g/cm³ at 300 °C and 500 MPa. However, when the temperature exceeded 300 °C, the higher temperature caused a violent reaction of powders to be squeezed out from the die. The increased evaporation rate of the liquid phase would also result in the insufficient wetting effect and dissolution process during the particle rearrangement process, leading to the fragment of the sample and slightly reduced density. Likewise, too large pressure exceeding 500 MPa could induce cracks after the removal of pressure, because of the increased risk of delamination and capping [40]. By contrast, prolonging the holding time showed a relatively weak impact on the final density (Fig. 4(c)). Thus, the CSP using 5 M NaOH solution with 40 wt% at 300 °C under 500 MPa for 30 min was determined as the optimal condition, which is much milder compared to the SPS.
The reason for such low densification temperature was first investigated by extracting the sintering activation energy using the Murray’s equation [41,42] as Eqs. (2) and (3) (the real density of the SPS-densified EMT was taken as the theoretical density of bulk NS-glass without considering the negligible influence of NaOH):

\[
\ln(1 - \rho) = -\frac{3\sigma}{4\eta}t + c \quad (2)
\]

\[
\eta = \frac{T}{K}\exp\left(\frac{E}{RT}\right) \quad (3)
\]

where \(\rho\) is the relative density, \(\sigma\) is the pressure (Pa), \(\eta\) is the viscosity (Pa·s), \(t\) is the time (s), \(c\) is the integration constant, \(E\) is the activation energy, \(K\) is the chemical equilibrium constant, \(T\) is the absolute temperature, and \(R\) is the gas constant. The slope of the \(\ln(1 - \rho)\) vs. \(t\) curve is followed to investigate the viscosity at a certain temperature. It can be observed that high sintering temperatures cause lower viscosity values, which were around \(4.53 \times 10^{11}\) Pa·s at 300 °C and \(2.28 \times 10^{12}\) Pa·s at 150 °C (Fig. 4(e)). The calculated values were similar to those found in hydrothermal hot pressing (HHP)-densified borosilicate glass [43] and CSP-densified soda-lime glass [28]. Furthermore, the activation energy of 27.13 kJ/mol (Fig. 4(f)) was obtained according to the linear dependence \((R^2 = 0.9901, \text{ wherein} R^2\text{ represents the “coefficient of determination” in the liner fitting})\) of the calculated viscosities to temperature, which is much lower than that of the HHP-densified aluminosilicate glass (around 300 kJ/mol) [44–46]. However, it is worth noting that, the densification with abnormally ultra-low activation energy should be ascribed to not only the viscous flow but also a complicated process involving the dissolution–precipitation, which leads to the grain growth under the chemical reaction effect. The chemical reaction was confirmed by comparing the X-ray spectroscopy (XPS) between various samples, as shown in Fig. 5 and Fig. S4 in the ESM. The high-resolution XPS spectra show that Si and Al peaks for the NS-glass shift to lower binding energy (Figs. 5(a) and 5(b)), which can be ascribed to the decreased covalent nature of the Si–O bond and Al–O bond within the structure by alkali etching [47,48]. O peaks in the C-EMT, DP-glass, and DW-glass show almost the same binding energy (531.6 eV) (Fig. 5(c)), which can be assigned to the co-existence of Si–O–Si (532 eV) and NaAlO (531 eV) structures in the aluminosilicate [49,50]. While the O peak in the NS-glass also shifts to lower binding energy (531.1 eV), probably owing to the increased ratio of NaAlO formed by the chemical reaction. In addition, the incorporation of Na in the matrix enhanced the ionic bonding force of Na–O [51], leading to the higher binding energy shift of the Na.
Fig. 5 High-resolution XPS spectra for (a) Al 2p, (b) Si 2p, (c) O 1s, and (d) Na 1s of C-EMT, DP-glass, DW-glass, and NS-glass (300 °C, 500 MPa, 5 M, 30 min).

peak in the NS-glass (Fig. 5(d)). Thus, the change in the chemical environment has clearly proved the chemical reaction between the powder and the solution, which could enhance the dissolution–precipitation process in the CSP.

Based on the above discussion, the schematic for the reaction mechanism between NaOH and C-EMT was proposed in Fig. S5 in the ESM. The framework of zeolite usually consists of SiO₄ and AlO₄ linked by O atoms. In a high concentration of alkaline solution within the CSP chamber, the highly hydrophilic nano-size C-EMT can be well dissolved, and the framework of zeolite would be damaged [52]. Specifically, the Si–O–Si bond tends to be easily attacked on the effect of OH⁻ while Si in the framework of zeolite can be extracted [48]. The Si–O–Al bond is frequently relatively stable under alkaline environment due to the negative charge of AlO₄⁻, which defends the Al from the attack of OH⁻. However, the rupture of the Si–Al–O bond would also probably occur during the CSP under the synergistic effect of the increased pressure and temperature [50]. Eventually, the uninterrupted chemical reaction promoted the densification process in such a short time with relatively low activation energy.

Thanks to the high density of cold-sintered NS-glass, the sample shows highly translucent in the visible-light spectrum (Fig. 6(b)). In comparison, the ZC pellet prepared from the EMT powder is completely opaque (Fig. 6(a)), indicating the successful strategy by using C-EMT here. It can be seen that, there is an abrupt change in the transmittance between 720 and 820 nm in all the cold-sintered glass bulk, which can also be observed in the Mn-doped aluminosilicate glass prepared by the SPS [53,54]. Considering the structure of sodium aluminosilicate glass [55,56], we deduced that the abnormal absorption could be ascribed to the changes in the Si–O–Si structure, which is reflected by the asymmetric stretching mode shifting (around 1000 cm⁻¹) in both DW-glass and DP-glass (Fig. 6(c)). Besides, the modification of Al pairs (Al–O–(Si–O)₂–Al) could also play a role [57]. Moreover, the bridging oxygen bonds could be broken into non-bridging silicon–oxygen groups by the chemical reaction of the C-EMT with the NaOH solution [58,59], which is evidenced by the presence of non-bridging silicon oxygen groups around 890 cm⁻¹ in the FTIR spectrum [60] (Fig. 6(c)). This kind of damage to the structure of glass also could cause the deterioration of transmittance.

As a result, the in-line transmittance of the NS-glass could just reach 18% at most in the visible-light spectra, which is still much lower compared with that of the aluminosilicate glass sintered by the conventional SPS [19]. We deduce that at least two factors could contribute to this difference in the transmittance. One is
that the density of the CSP-prepared glass is still not high enough, since the residual pores are highly detrimental to the transmittance. The other factor is related to the increased Na ratio derived from the chemical reaction between the powder and the NaOH solution, leading to the increased content of the secondary scattering phase, which has been proved by the above-mentioned EDS mappings. To prove this, the NS-glass sintered via adding the NaOH solution with various concentrations was prepared for comparison. It is found that adding a higher concentration (6 M) of the NaOH solution not only inversely deteriorated the transmittance of glass, but also made the glass more brittle, leading to a broken pellet after the CSP (Fig. 6(a)). Meanwhile, introducing a lower concentration (4 M) of the liquid aids gave rise to lower density (2.27 g/cm$^3$), which is also detrimental to the transmittance (Figs. 6(a) and 6(b)).

For realistic application, the mechanical properties of the cold-sintered aluminosilicate glass have to be evaluated, which is compared with those of the glass prepared by the SPS, as shown in Table 1 [31]. The CSP-prepared sample shows a decreased Young’s modulus, which can be attributed to the residual pores and the soft NaOH phase left in the matrix. As a result, the Vickers hardness of the NS-glass is also reduced by an almost identical extent with respect to the Young’s modulus (38.6% vs. 37.3%), due to the lack of grain boundary effect in the glass materials. In fact, although the hardness of the CSP-prepared glass is inferior to the SPS-prepared one, the value remains high enough for most applications. In contrast, the flexural strength has more than 60% reduction for the CSP-prepared glass, probably due to the fatal pores in the glass matrix. Similar to many reported ceramics densified by CSP, the mechanical properties of the CSP-prepared aluminosilicate glass are generally inferior to the bulks densified by the conventional methods, which could be enhanced if a further improved density can be realized.

| Sintering method | Density (g/cm$^3$) | Young’s modulus (GPa) | Flexural strength (MPa) | Vickers hardness (GPa) | Poisson’s ratio | Fracture toughness (MPa·m$^{1/2}$) |
|------------------|-------------------|----------------------|------------------------|-----------------------|----------------|-------------------------------|
| SPS              | 2.52              | 90                   | 88                     | 7                     | —              | —                             |
| CSP              | 2.41              | 56.4                 | 35                     | 4.3                   | 0.257          | 0.507                         |

*The relevant data are obtained from Ref. [31].

4 Conclusions

In conclusion, the translucent glass was prepared by the CSP at an ultra-low temperature of 300 °C for the first time. The EMT-type zeolite-derived nanoparticles

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can be densified to 2.41 g/cm³ via adding the NaOH solution, while the direct densification of the EMT powder only gave rise to an opaque compact after the CSP. The optimal sintering conditions were determined as 300 °C and 500 MPa by using 5 M NaOH solution as liquid aids, resulting in the highest in-line transmittance of 25% at 900 nm. After CSP, apparent grain growth was observed in the obtained NS-glass. Due to the homogeneous chemical reaction between C-EMT powder and NaOH solution, the consolidation was greatly accelerated by the dissolution–precipitation process at very low temperatures with the estimated activation energy of only 27.13 kJ/mol. In addition, too high and too low concentrations of the NaOH solution can both result in the deterioration of transmittance, indicating the importance of the liquid aids in the CSP. Finally, the CSP-prepared glass shows hardness high enough for common optical applications, although the flexural strength is much lower than the value of the sample prepared by the SPS.

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Declaration of competing interest

The authors have no competing interests to declare that are relevant to the content of this article.

Electronic Supplementary Material

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