Available online at www.sciencedirect.com

ScienceDirect

ELSEVIER

Journal of the European Ceramic Society 27 (2007) 4767–4772

www.elsevier.com/locate/jeurceramsoc

Low-temperature synthesis/densification and properties of Si_2N_2O prepared with Li_2O additive

Qingfeng Tong[a], Jingyang Wang[a], Zhongping Li[b], Yanchun Zhou[a,*]

[a] *Shenyang National Laboratory for Materials Science, Institute of Metal Research, Chinese Academy of Sciences, Shenyang 110016, PR China*

[b] *National Key Laboratory of Advanced Functional Composite Materials, Beijing 100076, PR China*

Received 27 December 2006; received in revised form 3 April 2007; accepted 14 April 2007
Available online 8 June 2007

Abstract

Dense Si_2N_2O was successfully synthesized using 2 mol% Li_2O as an additive by a hot-pressing method at 1500 °C. Compared to other metal oxide additives, Li_2O can significantly decrease the sintering temperature of Si_2N_2O, which is ascribed to the lower melting point of Li_2O–SiO_2 and the formation of less viscous liquid phase. Increasing Li_2O content has no apparent influence on the mechanical and dielectric properties of dense Si_2N_2O, which is due to the easy evaporation of Li_2O at sintering temperature. The mechanical properties of Si_2N_2O with Li_2O additive are comparable to those of Si_2N_2O synthesized with other additives. The as-prepared bulk Si_2N_2O with 2 mol% Li_2O additive exhibits both low dielectric constant (6.17 at 1 MHz) and loss tangent (0.0008 at 1 MHz) and combines good mechanical performance, indicating it is a potential high-temperature structural/functional material.

Keywords: Si_2N_2O; Sintering; Mechanical properties; Dielectric properties

1. Introduction

Silicon oxynitride (Si_2N_2O) is a unique compound in the SiO_2–Si_3N_4 system and exhibits many intrinsic properties which make it a promising high-temperature structural ceramic material.[1–6] It can keep excellent oxidation resistance in air up to 1600 °C and high flexural strength up to 1400 °C without degradation.[1,3–5] In addition, it possesses very low theoretic density (2.81 g/cm^3), high hardness (H_V: 17–22 GPa),[2] low thermal expansion coefficient (3.5×10^{-6} K^{-1}),[2] good thermal shock resistance and high thermodynamic stability temperature (about 1800 °C).[6]

Recently, based on ab initio calculations, Ching[7] predicted that Si_2N_2O possessed low dielectric constant, which is one of the most important physical properties for insulators. The good dielectric property of Si_2N_2O is mainly due to its bonding properties and special electronic structure which results in large band gap. However, there is no experimental result to test this conclusion mainly due to the fact that the synthesis of bulk Si_2N_2O is difficult.

The reaction sintering process of Si_2N_2O is similar to the sintering of Si_3N_4, i.e. both materials have strongly covalent bonds and low diffusion coefficient, requiring high sintering temperatures. This usually implies that oxide additives are needed to form a liquid phase with a eutectic melting point low enough to permit sintering without excessive dissociation. However, a high sintering temperature (≥1700 °C) and long holding time are still required even when various types of metal oxides (MgO, Al_2O_3, ZrO_2 and Ln_2O_3, where Ln represents Nd, Sm, Gd, Dy, Er, Yb, Y and Ce) additives are employed.[1–6] Compared to the aforementioned metal oxides additives, Li_2O is a promising additive owing to the lower melting point of Li_2O–SiO_2 and the formation of less viscous liquid phase. In the cases of sintering Si_3N_4 with $LiYO_2$ additive and AlN with $Li_2O + CaO + Y_2O_3$ additive, high density was obtained at a relatively low temperature because Li_2O-rich liquid phase could enhance the dissolution of reactants, accelerate the solution-diffusion–precipitation process, and facilitate the filling of pores.[8,9] In addition, the easy evaporation of Li_2O results in the low content of residual Li in as-sintered sam-

---

* Corresponding author. Tel.: +86 24 23971765; fax: +86 24 23891320.
E-mail address: yczhou@imr.ac.cn (Y. Zhou).

0955-2219/$ – see front matter © 2007 Elsevier Ltd. All rights reserved.
doi:10.1016/j.jeurceramsoc.2007.04.004

ples, which will be beneficial for high-temperature mechanical performance.

Due to easy evaporation of Li_2O at high temperature and enhanced sintering of Si_3N_4 and AlN,[8–10] Li_2O was selected as a sintering additive to prepare bulk Si_2N_2O in this work. The effect of Li_2O on the synthesis/sintering and densification of Si_2N_2O was investigated. The mechanical and dielectric properties of as-prepared bulk samples were measured. The results obtained in this paper are beneficial for the understanding of synthesis/sintering and properties of Si_2N_2O and for the promotion of Si_2N_2O material as a candidate for high-temperature structural/functional material.

2. Experimental procedures

Si_3N_4 powders (average particle size <0.36 μm, oxygen and total metallic impurity contents were 1.5 wt.% and less than 350 ppm, respectively, Sinoma Advanced Materials Co., Ltd., China) and SiO_2 powders (99.99% pure, Sinopharm Group Chemical Reagent Co., Ltd., China) were used as starting materials. To obtain an equimolar mixture of Si_3N_4 and SiO_2, the amount of Si_3N_4 was increased to compensate for its surface oxygen content. Li_2CO_3 powders (99% pure, Sinopharm Group Chemical Reagent Co., Ltd., China) as sintering aids were added to the equimolar Si_3N_4/SiO_2 mixture at 0.5–5 mol%. The mixture was ball-milled in agate jars using Si_3N_4 balls with methanol medium for 10 h. After being dried and sieved, the powders were put into high-strength graphite die whose inner surface was presprayed with a layer of BN and then cold-pressed at 5 MPa. Thereafter, the green body together with the die was heated in a nitrogen atmosphere at a heating rate of 10 °C/min and hot pressed under a pressure of 30 MPa at the temperatures of 1400, 1450, 1500 and 1550 °C for 1 h, respectively.

The density of as-sintered samples was measured by the Archimedes method. The XRD data of sintered bodies were collected using a step-scanning diffractometer with Cu Kα radiation (Rigaku D/max-2400, Japan). The quantitative phase analyses were performed using Rietveld method employing the DBWS code in the Cerius^2 computational program for materials research (Molecular Simulation Inc., USA).[11] The intensity is represented by

$$I_{\mathrm{Rietveld}}(2\theta) = b(2\theta) + S\sum_{K} L_K |F_K|^2 \phi(2\theta_i - 2\theta_K) P_K A_K \quad (1)$$

where b (2θ) is the background intensity; S the scale factor; L_K contains the Lorentz polarization and multiplicity factors; φ the profile function; P_K the preferred orientation function; A_K the absorption factor; F_K is the structure factor. The index K represents Miller indices for the Bragg reflections. In the Rietveld method, the mass fraction of a phase q, W_q is given by[12,13]

$$W_{\mathrm{q}} = \frac{S_{\mathrm{q}} M_{\mathrm{q}} V_{\mathrm{q}}}{\sum S_i M_i V_i} \quad (2)$$

where S is the Rietveld scale factor for the phase q; M the molar mass; V is the volume of the unit cell. The advantages of the Rietveld method include, viz. the calibration constants are computed rather than measured by experiments; all the peaks in the pattern play a part in the analysis; the use of a continuous fitting function provides a much improved background fit, and finally, the effects of preferred orientation and extinction are reduced.

Three-point flexural strength was determined at room temperature using rectangular bars (3 mm × 4 mm × 40 mm) in a universal testing machine at a crosshead speed of 0.5 mm/min. The Vickers hardness was measured at a load of 10 N with a dwell time of 15 s. Fracture toughness (K_{IC}) was determined by an indentation technique with a load of 49 N and a dwell time of 15 s. The fracture toughness was calculated using the equation proposed by Anstis et al.[14]

$$K_{\mathrm{IC}} = 0.016\left(\frac{E}{H}\right)^{1/2}\left(\frac{P}{C^{3/2}}\right) \quad (3)$$

$$H = \frac{P}{2a^2} \quad (4)$$

where E is the Young's modulus, P the applied indentation load, C the surface radial cracks length, H the Vickers hardness and a is the indentation half-diagonal length.

The Young's modulus and shear modulus at room temperature were evaluated in a RFDA-HTVP1750-C testing machine (IMCE, Diepenbeek, Belgium). The samples used were rectangular bars of 3 mm × 15 mm × 40 mm in size. The Young's modulus and shear modulus can be calculated according to the ASTM standard E 1259-94:

$$E = 0.9465\left[\frac{m f_{\mathrm{f}}^2}{b}\right]\left[\frac{L^3}{t^3}\right] T_1 \quad (5)$$

$$G = \frac{4Lm f_{\mathrm{t}}^2}{bt}\frac{B}{(1+A)} \quad (6)$$

where m is the mass of the bar; b, L, and t the width, length, and thickness of the bar, respectively; f_{f} the fundamental flexural resonant frequency of the bar; T_1 is the correction factor for fundamental flexural mode to account for factors such as the finite thickness of bar, and the Poisson's ratio. B and A are correcting factors depending on the width–thickness ratio.

The dielectric constant and loss tangent were determined using 4294A Impedance Analyzer and 16451B Dielectric Test Fixture (Agilent, California, USA). The frequency ranged from 40 Hz to 20 MHz. The microstructures of bulk samples were observed in a SUPRA 35 scanning electron microscope (LEO, Oberkochen, Germany). Before SEM observation, the sample was etched in molten NaOH for 30 s.

Inductively coupled plasma-atomic emission spectrometry (ICP-AES, Thermo Electron Corporation, Massachusetts, USA) was used to quantitatively determine the residual Li content in as-prepared samples.

3. Results and discussion

3.1. Sintering of Si_2N_2O with Li_2O addition

In the presence of a liquid phase obtained by the addition of metal oxide (Me_xO_y), the formation reaction of Si_2N_2O can be

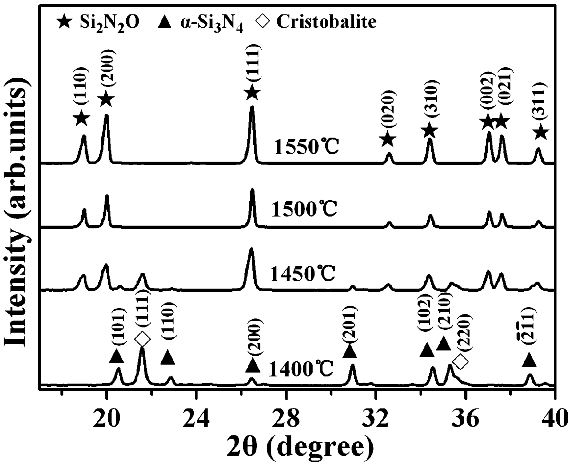

Fig. 1. X-ray diffraction patterns of as-sintered samples with 2 mol% Li_2O content in starting materials vs. synthesis/sintering temperature for 1 h holding time.

descried as the following reaction[3–5]:

$$Si_3N_4 + SiO_2 \xrightarrow{Me_xO_y} 2Si_2N_2O \quad (7)$$

The synthesis/sintering process of Si_2N_2O involves a liquid phase sintering mechanism. A liquid phase is formed by reaction of the metal oxide additives and SiO_2. And then Si_2N_2O is formed through a process whereby Si_3N_4 is dissolved and reacted with the SiO_2 component of the liquid phase. Generally speaking, the type and amount of metal oxide additives determine the formation temperature, the quantity and viscosity of the liquid phase. They also determine the temperature at which Si_2N_2O forms and densification commences and its rate during sintering.[2–5] Compared to other additives,[1–5] it is expected that Li_2O can lower the synthesis and sintering temperature of Si_2N_2O.

Fig. 1 shows the phase evolution of sintered bodies with 2 mol% Li_2O additive after heating at 1400–1550 °C. The content of Si_2N_2O was calculated using the Rietveld method (shown in Fig. 2). It can be seen that the starting temperature to form Si_2N_2O ranges from 1400 to 1450 °C. At 1500 °C, the fraction of Si_2N_2O reaches the maximum and the density of as-prepared sample is close to the theoretical value. Further increase of sintering temperature does not improve the content of Si_2N_2O and density of the resulting specimen.

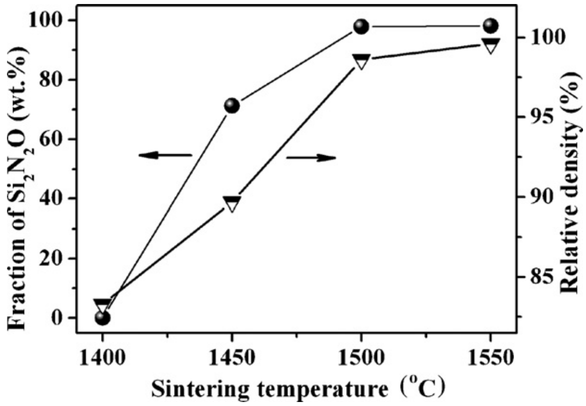

Fig. 2. The relative density and the fraction of Si_2N_2O in as-sintered samples with 2 mol% Li_2O in starting materials vs. sintering temperature for 1 h holding time.

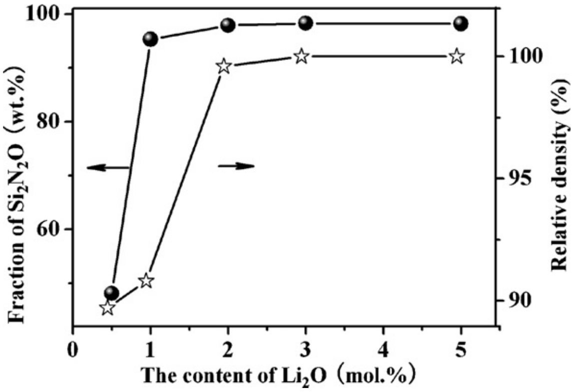

Fig. 3. The relative density and fraction of Si_2N_2O in samples sintered at 1550 °C for 1 h vs. Li_2O content in starting materials.

Fig. 3 shows the effect of Li_2O content in starting materials on the relative density and fraction of Si_2N_2O in as-sintered samples at 1550 °C. Both the relative density and the fraction of Si_2N_2O increase with increasing Li_2O contents. When 2 mol% Li_2O is added, almost fully dense sintered body with 98 wt.% Si_2N_2O is obtained. And with further increasing Li_2O content in the starting materials, the density of as-prepared sample and fraction of Si_2N_2O are not improved.

Based on the above results, it can be concluded: (1) the addition of 2 mol% Li_2O is enough to prepare the dense and pure Si_2N_2O; (2) Li_2O significantly lowers the synthesis and sintering temperature of Si_2N_2O, which is attributed to the lower melting point of Li_2O–SiO_2 and the formation of a less viscous liquid phase at the sintering temperature. The eutectic point of Li_2O/SiO_2 is about 1255 °C,[8–10] which is much lower than that of other metal oxide additives/SiO_2 (>1540 °C).[15–17] The presence of Li_2O will lead to a less viscous liquid phase. This liquid phase improves the wetting and dissolution of Si_3N_4, accelerates the precipitation and growth of Si_2N_2O grains, and results in the rapid densification of Si_2N_2O at a relatively low temperature.

3.2. Mechanical properties of as-prepared Si_2N_2O

The microstructure and properties of Si_2N_2O with Li_2O additive are studied and compared to those employing other metal oxides in order to investigate the effect of Li_2O. Fig. 4 shows the microstructure of Si_2N_2O synthesized at 1550 °C using 2 mol% Li_2O as a sintering additive. It displays similar morphology to microstructures of Si_2N_2O prepared using other metal oxide as sintering additives.[1–5] Si_2N_2O grains exhibit plate-like and rod-like shape with the average size of about 1–3 μm.

Table 1 exhibits relative densities, mechanical properties and residual Li content in as-prepared samples. When the Li_2O content in the starting material is above 2 mol%, the influence of the Li_2O content on the mechanical properties of the as-prepared sample is not pronounced, which is attributed to the easy evaporation of Li_2O during firing. In the starting mate-

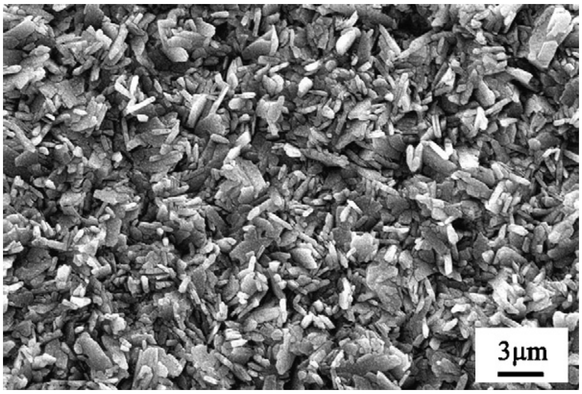

Fig. 4. Scanning electron micrograph of a Si_2N_2O sample sintered at 1550 °C for 1 h. The sample was etched by molten NaOH.

rial, the initial contents of Li element are designed to be 0.14, 0.21 and 0.35 wt.% Li (equivalent to 2, 3 and 5 mol% Li_2O content, respectively). After the green compacts are sintered at 1550 °C for 1 h, the residual Li contents drop to 0.06, 0.07 and 0.09 wt.%, respectively. In order to further lower the Li content, the as-sintered sample with 2 mol% Li_2O in starting material was annealled at 1550 °C for 2 h. Its residual Li content were measured to be 0.05 wt.%. The vapor pressure of Li_2O reaches about 5 Pa at 1550 °C.[9] Therefore, the Li_2O is easily volatilized from the specimen at the firing temperature and the residual Li_2O content remains low in the as-prepared samples. In the case of pressureless sintering of AlN and Si_3N_4, the residual Li_2O content in as-sintered samples was measured to be 1/100 and 1/20 of starting materials, respectively.[8–10] In the present work, more amounts of Li are retained even when the as-prepared sample is annealled for 2 h, which is due to the lower sintering and annealling temperature, shorter holding time, higher relative density and less diffusion channels. It can also be seen in Table 1 that the residual Li slightly increases with the increasing of Li contents in the starting materials. Since the residual Li may lower the softening temperature of grain boundary and is deleterious to the high temperature properties,[18] 2 mol% Li_2O content is assumed to be an appropriate value to prepare dense and pure Si_2N_2O and annealling is helpful to further lower the residual Li content.

Table 2 compares the properties of dense Si_2N_2O with various metal oxide additives.[1–5] The result demonstrates that the mechanical properties of dense Si_2N_2O with Li_2O additive are comparable to those of Si_2N_2O synthesized by other method and additives. But the sintering temperature is greatly decreased and the firing time is shortened.

3.3. *Dielectric properties of as-prepared Si_2N_2O*

Besides mechanical properties, dielectric constant is the important performance parameter of structural/functional material. Figs. 5 and 6 show dielectric constant ε' and loss tangent tg δ of as-prepared Si_2N_2O with various Li_2O content, respectively. It can be seen that the dielectric constant decreases continuously with increasing frequency. The loss tangent is below 0.006 in the frequency range of 40 Hz to 20 MHz. When the Li_2O content in starting materials is above 2 mol%, the effect of Li_2O content on the dielectric constant and loss tangent is not pronounced.

Generally, the dielectric constant of materials decreases while loss tangent increases with the improvement of relative density, respectively.[19] Therefore, Si_2N_2O with 1 mol% Li_2O shows lower dielectric constant than that with above 1 mol% Li_2O. However, for the as-sintered sample with 1 mol% Li_2O, 10 vol.% pores has no obvious influence on its loss tangent.

Table 1
Properties of sintered Si_2N_2O with various Li_2O content in starting materials

Properties	The content of Li_2O (mol%)			
	1	2	3	5
Relative density (%)	90.7	99.6	100	100
Bending strength (MPa)	162 ± 15	513 ± 17	502 ± 15	497 ± 20
K_{IC} (MPa m$^{1/2}$)	1.8 ± 0.1	3.3 ± 0.3	2.9 ± 0.2	2.8 ± 0.3
E (GPa)	192	229	229	228
G (GPa)	85	100	99	98
Hardness (GPa)	9.5 ± 0.4	17.1 ± 0.4	16.8 ± 0.4	16.6 ± 0.8
The Li content in starting material (wt.%)	0.07	0.14	0.21	0.35
Residual Li content in as-sintered sample (wt.%)	–	0.06	0.07	0.09
Residual Li content after annealling for 2 h (wt.%)	–	0.05	–	–

Table 2
Comparison of properties of Si_2N_2O with various metal oxide additives

Properties	2 mol% Li_2O	2 mol% (MgO)	20 mol% (Al_2O_3)	1.5 mol% (Y_2O_3)	1.5 mol% (Sm_2O_3)	HIP (no additive)
ρ (g/cm^3)	2.81	2.82	2.86	2.87	2.90	2.84
Fraction of Si_2N_2O (wt.%)	97.9	98	98	91	99	91.1
σ_b (MPa)	513	493	480	655	638	490
K_{IC} (MPa m$^{1/2}$)	3.3	2.5	2.4	5.1	3.2	3.1
Sintering temperature (°C)	1550	1700	1700	1700	1700	1950
Holding time (h)	1	2	2	2	2	4

Table 3
Selected properties of several structural/functional ceramics

Properties	Si_2N_2O	Si_3N_4	Al_2O_3	AlN	Hexagonal BN	Fused SiO_2
ρ (g/cm^3)	2.81	3.18	3.98	3.2	2.27	2.2
E (GPa)	289	320	416	330	77.9	72
σ_b (MPa)	513	700	380	280	89.6	50
K_{IC} (MPa m$^{1/2}$)	3.3	4.5	2.7	2.6	–	1.0
H_V (GPa)	17	16.5	18	12	2 (Morse)	7
ε' (1 MHz)	6.17	9	9.9	9.4	5.16	3.9
tg δ (1 MHz)	0.0008	0.002	0.00023	0.002	0.0002	0.0003
CTE (10^{-6} °C^{-1})	3.3	3.2	8.2	4.3	6	0.75
References	This work, 4	20,21	20,22	20,23	20	20,24

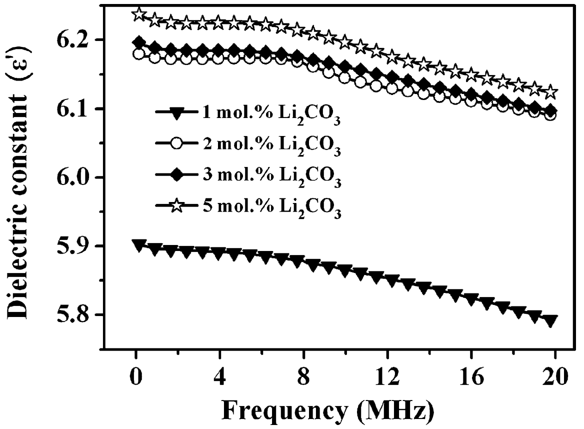

Fig. 5. The frequency dependence of dielectric constant of as-prepared Si_2N_2O vs. Li_2O content in starting materials.

The effect of Li_2O content in starting materials on dielectric properties can be ignored because of the volatilization of Li_2O during firing. This trend is similar to the influence of Li_2O content in starting materials on mechanical properties of Si_2N_2O.

Table 3 lists some properties of high-temperature structural ceramics with low dielectric constant and loss tangent. The dielectric constant of Si_2N_2O is lower by about 3–4 than that of Si_3N_4, Al_2O_3 and AlN, and its flexural strength and fracture toughness obviously excels Al_2O_3 and AlN. In comparison with BN, the dielectric properties of Si_2N_2O are slightly inferior while its mechanical properties are much better. Fused SiO_2 receives much attention due to its low coefficient of thermal expansion, excellent thermal shock resistance and good dielectric properties,[25] however, fused SiO_2 as a high-temperature structural ceramic has some shortcomings, such as its poor flexural strength and fracture toughness and the further degradation of mechanical properties resulting from softening and crystallization of amorphous SiO_2 at above 1100 °C.[26] Compared with fused SiO_2, Si_2N_2O possesses slightly inferior dielectric properties, however, its flexural strength and fracture toughness are about 10 and 3 times higher than that of fused SiO_2, respectively, and it can keep satisfied chemical stability up to 1750 °C.

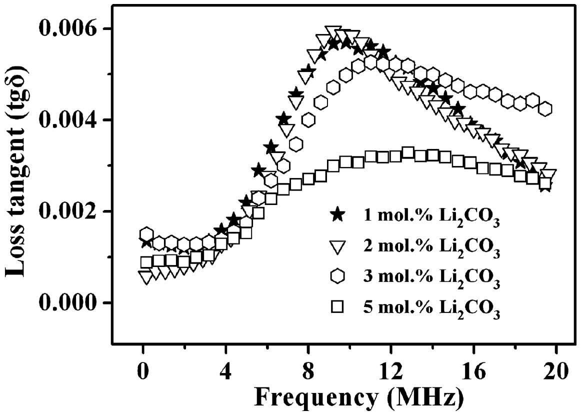

Fig. 6. The frequency dependence of loss tangent of as-prepared Si_2N_2O vs. various Li_2O content in starting materials.

4. Conclusions

(1) In comparison with other metal oxide sintering additives, Li_2O is more effective in synthesizing pure and dense Si_2N_2O. The synthesis/sintering temperature is greatly lowered and the firing time is shortened.
(2) Dense Si_2N_2O with Li_2O additive exhibits similar mechanical properties to those using other additives. The content of Li_2O in starting materials shows little influence on the mechanical and dielectric properties of Si_2N_2O due to the evaporation of most Li_2O at high temperatures.
(3) Si_2N_2O combines good mechanical and dielectric performance, indicating it is a promising candidate for high-temperature structural/functional material.

Acknowledgements

This work was supported by the National Outstanding Young Scientist Foundation of China under grant no. 59925208, Natural Sciences Foundation of China under grant nos. 50232040, 50072034, 90403027, 863 project, and High-tech Bureau of the Chinese Academy of Sciences.

References

1. Ohashi, M., Kanzaki, S. and Tabata, H., High-temperature flexural strength of hot-pressed silicon oxynitride ceramics. *J. Mater. Sci. Lett.*, 1988, **7**, 339–340.
2. Larker, R., Reaction sintering and properties of silicon oxynitride densified by hot isostatic pressing. *J. Am. Ceram. Soc.*, 1992, **75**, 62–66.

3. Ohashi, M., Kanzaki, S. and Tabata, H., Processing, mechanical properties and oxidation behavior of silicon oxynitride ceramics. *J. Am. Ceram. Soc.*, 1991, **74**, 109–114.
4. Ohashi, M., Kanzaki, S. and Tabata, H., Effect of additives on some properties silicon oxynitride ceramics. *J. Mater. Sci.*, 1991, **26**, 2608–2614.
5. Huang, Z. K., Greil, P. and Petzow, G., Formation of silicon oxynitride from Si_3N_4 and SiO_2 in the presence of Al_2O_3. *Ceram. Int.*, 1984, **10**, 14–17.
6. Rocabois, P., Chatillon, C. and Bernard, C., Thermodynamics of the Si–O–N system. II. stability of Si_2N_2O(s) by high-temperature mass spectrometric vaporization. *J. Am. Ceram. Soc.*, 1996, **79**, 1361–1365.
7. Ching, W. Y., Electronic structure and bonding of all crystalline phase in the silica–yttria–silicon nitride phase equilibrium diagram. *J. Am. Ceram. Soc.*, 2004, **87**, 1996–2013.
8. Matovic, B., Rixecker, G. and Aldinger, F., Pressureless sintering of silicon nitride with lithia and yttia. *J. Eur. Ceram. Soc.*, 2004, **24**, 3395–3398.
9. Matovic, B., Rixecker, G. and Aldinger, F., Densification of Si_3N_4 with $LiYO_2$ additive. *J. Am. Ceram. Soc.*, 2004, **87**, 546–549.
10. Watari, K., Valecillos, M. C., Brito, M. E., Toriyama, M. and Kanzaki, S., Densification and thermal conductivity of AlN doped with Y_2O_3, CaO and Li_2O. *J. Am. Ceram. Soc.*, 1996, **79**, 3103–3108.
11. Young, R. A., *The Rietveld Method*. Oxford University Press, 1993.
12. Wiles, D. B. and Young, R. A., A new computer program for Rietveld analysis of X-ray powder diffraction patterns. *J. Appl. Crystallogr.*, 1981, **14**, 149–151.
13. Hill, R. J. and Howard, C. J., Quantitative phase analysis from neutron powder diffraction data using the Rietveld method. *J. Appl. Crystallogr.*, 1987, **20**, 467–474.
14. Anstis, G. R., Chantikul, P., Lawn, B. R. and Marshall, D. B., A critical evaluation of indentation techniques for measuring fracture toughness. I. Direct crack measurements. *J. Am. Ceram. Soc.*, 1981, **64**, 533–538.
15. Staronka, A., Pham, H. and Rolin, M., Study of system SiO_2–Al_2O_3 by cooling curve method. *Rev. Int. Haules Temp. Refract.*, 1968, **5**, 111–115.
16. Kambayshi, S. and Kato, E., A thermodynamic study of (magnesium oxide + silicon dioxide) by mass spectrometry. *J. Chem. Thermodyn.*, 1984, **16**, 241–248.
17. Torpov, N. A., Some rare earth silicates. In *Trans. Int. Ceram. Congr. 7th*, 1960, pp. 435–442.
18. Ziegler, G., Heinrich, J. and Wotting, G., Review relationship between processing, microstructure and properties of dense and reaction-bonded silicon nitride. *J. Mater. Sci.*, 1987, **22**, 3041–3086.
19. Walton Jr, J. D., Reaction sintered silicon nitride for high temperature radome applications. *Ceram. Bull.*, 1974, **53**, 255–258.
20. Ceramic properties databases of the American Ceramic Society. Available from: http://www.matweb.com/search/SpecificMaterial.aspbassnum.
21. Hampshire, S., Engineering properties of nitrides. In *Engineered Materials Handbook*, *Vol. 4*, ed. S. J. Schneider Jr. ASM International, 1991, pp. 812–820.
22. Munro, R. G., Evaluated material properties for a sintered alpha-Al_2O_3. *J. Am. Ceram. Soc.*, 1997, **80**, 1919–1928.
23. Khor, K. A., Cheng, K. H., Yu, L. G. and Boey, F., Thermal conductivity and dielectric constant of spark plasma sintered aluminum nitride. *Mater. Sci. Eng. A*, 2003, **347**, 300–305.
24. Lyons, J. S. and Starr, T. L., Strength and toughness of slip-cast fused-silica composites. *J. Am. Ceram. Soc.*, 1994, **77**, 1673–1675.
25. Wen, G., Wu, G. L., Lei, T. Q., Zhou, Y. and Guo, Z. X., Co-enhanced SiO_2–BN ceramics for high-temperature dielectric applications. *J. Eur. Ceram. Soc.*, 2000, **20**, 1923–1928.
26. Bruckner, R., Properties and structure of vitreous silica. *J. Non-Cryst. Solids*, 1970, **5**, 123–175.