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Synthesis and characterization of leucite ceramics using sol–gel derived molecular precursors

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Monophasic leucite ($KAlSi_2O_6$) was synthesized from a K–Al–Si–O gel precursor at 950 °C.

Artificial bone materials used in dentistry and orthopedy are of considerable current interest.[1] Dental biomaterials should be biocompatible and simulate the characteristics of teeth.[2] Leucite ($KAlSi_2O_6$) provides such properties, as it can be manufactured in ceramic–metal or all-ceramic prosthetic structures.[3] Leucite dental ceramic is widely used in porcelain-fused-to-metal (PFM) structures because it is compatible with most of the metals used in prosthodontics.[4–6]

Leucite can be prepared from active alumosilicates such as metakaolin,[7] kaolin,[8] fly ash *etc*.[9–12] Leucite crystallizes in association with a kalsilite ($KAlSiO_4$) phase, and kalsilite crystallizes along with or before leucite as a metastable phase.[5,7,13] This can be due to the fact that kalsilite has a lower activation energy than leucite.[13] In some cases, this intermediate changes to leucite at higher temperatures or upon prolonged thermal treatment.[5]

It is well known that sol–gel methods can provide the syntheses of oxide materials at lower temperatures.[5,14–16] In this study, we used an inexpensive and environmentally benign aqueous sol–gel technique for the preparation of $KAlSi_2O_6$ from analytical grade reagents.†

The phase composition of the products synthesized at different temperatures was determined by XRD analysis.‡ These results showed that an amorphous material was obtained after the heating of K–Al–Si–O gel precursors at 700 °C for 2 h. However, the situation was different with increasing the annealing temperature up to 950 °C (Figure 1). The XRD pattern of a sample synthesized at 950 °C shows the formation of a crystalline leucite phase. However, a small impurity peak at $2\theta \approx 28.64°$ confirms the formation of a kalsilite phase. On the other hand, this peak is much weaker in comparison with the main leucite reflections and indicates mainly the incomplete crystallization of leucite, when presumptively kalsilite reacts with the residual SiO_2. The monophasic leucite was formed after increasing the annealing temperature to 1000 °C using 24 h treatment duration. Figure 1 shows that the single phase leucite (reference data PDF [038-1423]) with strong and distinct peaks at $2\theta \approx 25.84$ and 27.30° was formed and no traces of kalsilite were detected.

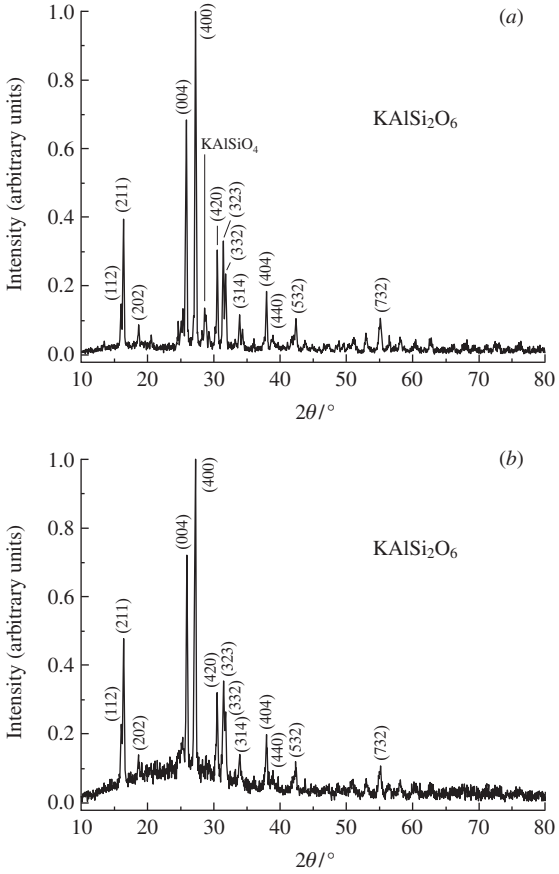

Figure 1 XRD patterns of K–Al–Si–O gel precursors annealed at (*a*) 950 °C for 5 h and (*b*) 1000 °C for 24 h.

Figure 2 demonstrates the IR spectra of the ceramic powders obtained after calcination at 950 °C for 5 h and at 1000 °C for 24 h. The broad bands located between 3500 and 3350 cm^{-1} and peaks in the region of 1620–1640 cm^{-1} correspond to adsorbed atmospheric moisture. The appearance of small peaks at ~2400 cm^{-1} is due to adsorbed CO_2. In both spectra, three peaks in the range of 1300–500 cm^{-1}, characteristic of the leucite structure, are

† First, fumed silicon dioxide (SiO_2) was dissolved in 100 ml of distilled water with continuous stirring for 24 h at 65 °C. Potassium hydroxide and aluminum nitrate nonahydrate were dissolved separately in small amounts of distilled water. Both of the solutions were slowly added to the starting homogenized sol. Next, the prepared sol was stirred for 1 h at the same temperature. To prevent the crystallization of aluminum hydroxide during the gelation processing, 1,2-ethanediol (2 ml) was added as a complexing agent. The sol was concentrated by the slow evaporation of water at the same temperature in an open beaker. The resulting gels were dried in an oven for 24 h at 100 °C and then calcined at 700 °C for 5 h. Finally, after intermediate grinding, the ceramic materials were annealed in a corundum crucible at 750 to 1000 °C (the heating rate was 5 K min^{-1}).

‡ Powder X-ray diffraction analysis was carried out using a Rigaku MiniFlex II diffractometer with the Bragg–Brentano ($\theta/2\theta$) geometry. The data were collected in the range of 2θ angle from 10° to 80° at a step of 0.02° and integration time of 1 s using the Ni-filtered CuKα line. A scanning electron microscope (SEM, EVO 50 XVP) was used to study the surface morphology of the materials. The Fourier transform IR (FTIR) spectra were measured using a Perkin–Elmer FTIR Spectrum BX II spectrometer in the range of 4000–400 cm^{-1}.

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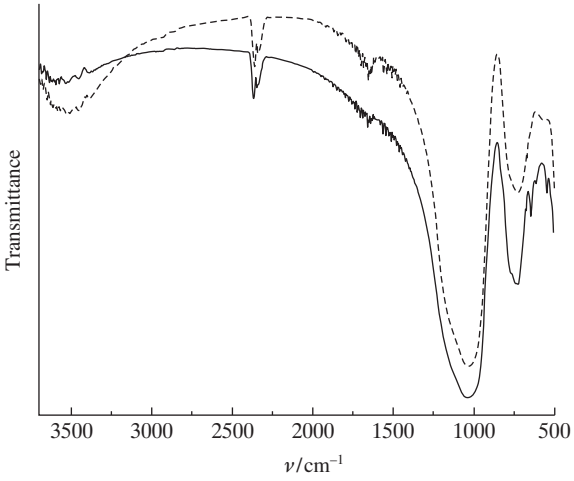

Figure 2 IR spectra of leucite ($KAlSi_2O_6$) after annealing at 950 °C for 5 h (dashed line) and 1000 °C for 24 h (solid line).

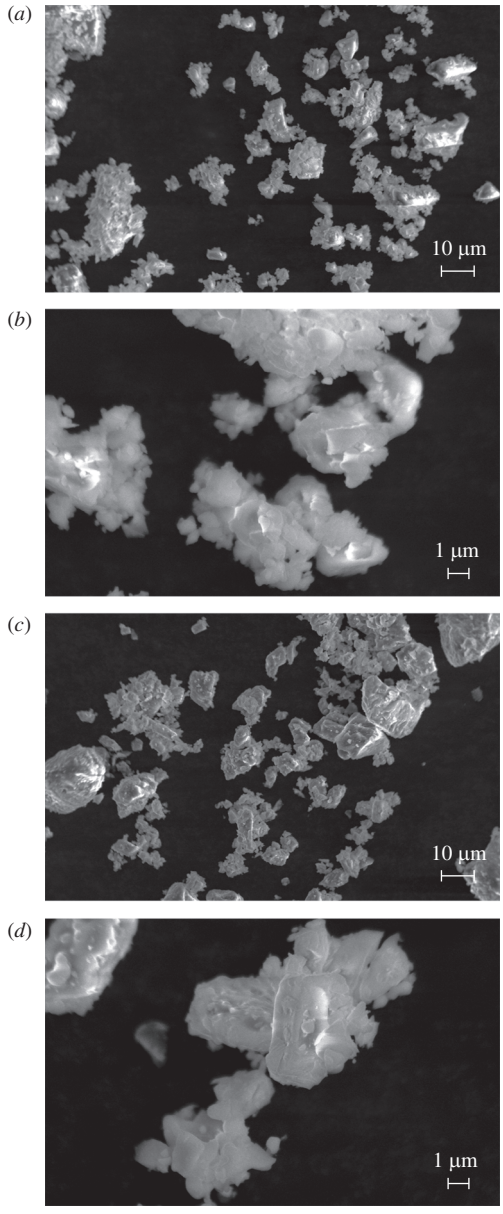

Figure 3 SEM micrographs of leucite ceramics synthesized at different temperatures: (*a*),(*b*) 950 °C and (*c*),(*d*) 1000 °C and obtained at different magnifications.

present. The most intense peak of Si–O–Si groups, located in the 1034–1010 cm^{-1} region, is due to asymmetric and symmetric stretching vibrations of the tetrahedral network.[17] The broad bands and peaks in the range of 850–630 cm^{-1} correspond to the Al–O stretching vibrations of AlO_x polyhedral units.[18]

The SEM images of the ceramics synthesized at 950 °C for 5 h and at 1000 °C for 24 h are shown in Figure 3. In both cases, crystalline particles with similar surface morphology were formed. Differently shaped particles of size 0.5–5 μm can be observed. The synthesis products consist of clustered grains made up of several tiny crystallites with a defined structure.[19] In addition, the $KAlSi_2O_6$ solids obtained at 1000 °C exhibit a slightly increased agglomeration of the crystallites.

In conclusion, leucite ceramic was synthesized from molecular precursors using environmentally benign sol–gel chemistry. The monophasic leucite free from a kalsilite impurity was formed at 1000 °C. Fine crystallites with an average size of 0.5–5 μm were obtained. Thus, a simple and economic synthesis of the monophasic dental material leucite has been elaborated.

References

1 C. P. Yoganand, V. Selvarajan, L. Lusvarghi, O. M. Goudouri, K. M. Paraskevopoulos and M. Rouabhia, *Mater. Sci. Eng.*, 2009, **C29**, 1759.
2 W. Holand and V. Rheinberger, *J. Mater. Sci.: Mater. Med.*, 2006, **17**, 1037.
3 X.-F. Song, L. Yin and Y.-G. Han, *J. Med. Eng. Phys.*, 2009, **31**, 214.
4 N. Xie, J. L. Bell and W. M. Kriven, *J. Am. Ceram. Soc.*, 2010, **93**, 2644.
5 Y. Zhang, J. Wu, P. Rao and M. Lü, *Mater. Lett*., 2006, **60**, 2819.
6 M. Kohoutkova, A. Klouzkova, P. Kostka and M. Mrazova, *J. Non-Cryst. Solids*, 2008, **354**, 741.
7 P. He, D. Jia, M. Wang and Y. Zhou, *Ceram. Int.*, 2011, **37**, 59.
8 S. Hashimoto, A. Yamaguchi, K. Fukuda and S. Zhang, *Mater. Res. Bull.*, 2005, **40**, 1577.
9 V. F. F. Barbosa and K. J. D. MacKenzie, *Mater. Lett.*, 2003, **57**, 1477.
10 M. J. Cattell, T. Chadwick and J. Knowles, *J. Dent. Mater.*, 2006, **22**, 925.
11 S. Hashimoto, F. Sato, S. Honda, H. Awaji and K. Fukuda, *J. Ceram. Soc. Jpn*., 2005, **113**, no. 1319, 488.
12 P. He, D. Jia, M. Wang and Y. Zhou, *Ceram. Int.*, 2010, **36**, 2395.
13 Y. Zhang, M. Lü, D. Chen and J. Wu, *Mater. Lett.*, 2007, **61**, 2978.
14 G. Peleckis, K. Tonsuaadu, T. Baubonyte and A. Kareiva, *J. Non-Cryst. Solids*, 2002, **311**, 250.
15 A. Fernandez-Jimenez, R. Vallepu, T. Terai, A. Palomo and K. Ikeda, *J. Non-Cryst. Solids*, 2006, **352**, 2061.
16 I. Bogdanoviciene, A. Jankeviciute, J. Pinkas, A. Beganskiene and A. Kareiva, *Mater. Res. Bull.*, 2008, **43**, 2998.
17 M. Handke, M. Sitarz, M. Rokita and E. Galuskin, *J. Mol. Struct.*, 2003, **651–653**, 39.
18 N. Dubnikova, E. Garskaite, J. Pinkas, P. Bezdicka, A. Beganskiene and A. Kareiva, *J. Sol-Gel Sci. Technol*., 2010, **55**, 213.
19 S. Sakirzanovas, L. Sun, C. Yan and A. Kareiva, *Mendeleev Commun.*, 2008, **18**, 251.

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