Some sialons prepared from nanopowders by hot pressing

I. Zalite¹, N. Zilinska¹, G. Kladler²

¹ Institute of Inorganic Chemistry of the Riga Technical University, 34 Miera Str., Salaspils, LV-2169, Latvia, ² ARC Seibersdorf research GmbH, A-2444, Seibersdorf, Austria

ilmars@nki.lv

Abstract. Investigation of fabrication of α-sialon and α-/β-sialon ceramics from nanopowders by hot pressing (1800 and 1910 °C) were carried out. Samples of sialons were made by using Si₃N₄-AlN nanopowders composites fabricated by the method of plasmachemical synthesis. The hardness of materials from nanopowders are of HV₁= 17,6-21,9 GPa, bending strength of σ₃ₚ= 520-800 MPa and fracture toughness up to K₁c= 7,1 MPa.m¹/₂, depending on composition and compacting temperature.

1. Introduction

Silicon nitride based ceramics, including sialons, due to their increased mechanical properties (hardness, mechanical strength, good corrosion and oxidation resistance at elevated temperature as well as bad wetting ability with metal alloys) are promising candidates for structural materials [1,2] for high-temperature application and production of ceramic matrix composites. A lot of investigations [3-5] have been made during last years on preparation of sialon ceramics, especially on the effect of microstructure of materials on their properties.

Sialon ceramics are being prepared from powders and therefore properties of ceramic materials depend to a large extent on the quality of the starting powders. The powder determines the processing, the sintering behaviour and the subsequent formation of the microstructure, which strongly influences many properties of the dense materials. The diffusion velocity and the length of diffusion way are of great importance for compacting of covalent compounds, for example, Si₃N₄ and sialons. The length of diffusion way can be decreased by using nanocomponents for production of compact material. As it is obvious from many investigations of compacting, the application of Si₃N₄ nanopowders enables to decrease the sintering temperature and to accelerate the sintering process, resulting in material with more fine-grained structure and modified properties.

One of the ways how to impact the microstructure of materials should be application of nanosized raw powders for production of ceramics. As it is obvious from many investigations of compacting [6], the application of Si₃N₄ nanopowders enables to decrease the sintering temperature and to accelerate the sintering process resulting in material with more fine-grained structure and modified properties.

The aim was to investigate the compacting process of sialon nanopowders of several compositions by hot pressing method as well as comparison of ceramics with β-Si₃N₄ materials obtained at the similar conditions from nanopowders.
2. Experimental

Samples of α-/β- and α-sialon of differing compositions were made by using of plasma-synthesized Si₃N₄-AlN nanocomposites containing 86 wt.% Si₃N₄, 12 wt.% AlN, 0.6 wt.% Si₆ecz, with the specific surface area (SSA) of 50 m²/g and average particle size (d₅₀) of 40 nm (micrograph of this nanopowder is given at Fig.1.) and small amount of AlN (H.C.Starck, grade B), Al₂O₃ (Alcoa Chemie GmbH, A16SG) and Y₂O₃ (H.C.Starck, grade C) powder additions for correcting of the composition. The specimen compositions used are in Table 1. The standard Si₃N₄-6 wt.% Y₂O₃-3 wt.% Al₂O₃ nanopowder was also used for comparison. Nanopowders were produced by the method of plaschemical synthesis, where the mixture of raw powders was evaporated in radio-frequency nitrogen plasma with the following condensation of reaction products as nanopowders from gas/vapour phase [7].

![Micrograph of Si₃N₄-AlN nanopowder.](image)

**Table 1.** Composition of sialon specimens, wt. %

| No. | Chemical composition, wt.% | Phase       |
|-----|----------------------------|-------------|
|     | Si₃N₄ | AIN | Al₂O₃ | Y₂O₃ |           |
| 1   | 90.4  | -   | 3.2   | 5.9  | β- Si₃N₄ |
| 2   | 85.3  | 9.2 | 2.1   | 3.4  | β- / α- sialon |
| 3   | 76.3  | 15.6| 0.2   | 7.9  | α- sialon |
| 4   | 74.4  | 13.2| -     | 12.4 | α- sialon |
| 5   | 72.1  | 16.3| 2.5   | 8.2  | α- sialon |

Simultaneously at the same conditions (for comparison of sintering process) separate sialon ceramics samples were obtained also from commercial powders: α-Si₃N₄ (UBE, SN-10E), AIN (H.C.Starck, Grade C), Al₂O₃ (Alcoa Chemie GmbH, A16SG) and Y₂O₃ (Nanophase).

All powders were homogenized in hexane for 15 h in a rotating polyethylene bottle with silicon nitride balls, afterwards treated for 2 hours in an ultrasonic bath. After the following drying at 80 °C the powder was sieved through a 200 µm mesh.

Hot pressing (1800 and 1910 °C) was used for compacting of materials. The temperature increasing rate was 10°/min, isothermal holding time – 2 hours, sintering medium – nitrogen.

Chemical composition of the nanopowders (N, Si₆ecz, Al) was determined by the chemical analysis. Phase composition of the sintered specimens was performed via X-ray diffractometry (XRD). The form and size of particles were determined by the transmitting electron microscope (TEM). The microstructure was observed using scanning electron microscopy (SEM) on fracture surface. Density of the sintered samples was determined by the Archimedes method. Hardness (load: 1 kg) and fracture toughness (load: 10 kg) were measured by the Vickers indentation technique.
3. Results and discussion

![Fig. 2. Microstructure of samples hot pressed at 1800 °C.]

As it can be seen from Fig. 2 the material structure depends on chemical composition: β-silicon nitride and β-sialon has typical needle-shaped structure. Due to this material possess high bending strength (Table 2.), also at high temperatures. Depending on composition α-sialons can have different structure of material, however the presence of needle-shaped crystals is less and therefore the bending strength is also lower, while the hardness is significantly higher. Grain size of materials obtained by hot pressing is comparatively high (0,5-1,0 µm), but the length of needle-shaped crystals is of 5-10 µm (this is more than in the case of sialon materials of similar composition obtained after sintering up to 1700 °C [8]).

Table 2. Properties of sialon ceramics prepared by hot pressing (N₂, 2 h)

| Sample | T, °C | ρ, g/cm³ | P₁₀₂₀, % | HV₁₀₂₀, GPa | σ₂₀, MPa | E₂₀, GPa | σ₁₀₀₀, MPa | E₁₀₀₀, GPa | K₁c, MPa·m⁻¹/₂ |
|--------|------|----------|----------|-------------|----------|---------|-------------|---------|-------------|
| 1      | 1800 | 3,17     | 0,2      | 17,0        | 690      | 235     | 550         | 135     | 4,4         |
| 1580   | 3,27 |          | 0,0      | 16,8        | 1150     | 310     | -           | -       | -           |
| 2      | 1800 | 3,20     | 0,6      | 20,0        | 715      | 215     | -           | -       | -           |
| 1910   | 3,22 |          | 0,0      | 17,6        | 760      | 335     | 755         | 181     | -           |
| 3      | 1800 | 3,26     | 0,2      | 21,1        | 535      | 250     | 221         | 140     | -           |
| 1910   | 3,26 |          | 0,0      | 20,2        | 620      | 330     | -           | -       | -           |
| 4      | 1800 | 3,33     | 0,1      | 21,9        | 345      | 245     | -           | -       | -           |
| 1910   | 3,32 |          | 0,0      | 20,8        | 520      | 340     | -           | -       | -           |
| 5      | 1910 | 3,24     | 0,0      | 21,4        | 680      | 250     | -           | -       | 7,1         |
With the increase of hot pressing temperature the needle-shaped structure becomes more evident and the bending strength increases but hardness decreases.

As it is shown from densification process of hot pressing (Fig. 3, \( \frac{dl}{dt} \)), kinetics of densification of nanopowders and industrial powders are significantly differing: in the case of nanopowders more significant is the starting stadium of compacting (at 1430-1550 °C), when the density grows due to the grain surface slipping.

Fig. 3. Process of the hot pressing of samples 5
(A- industrial powders, B- nanopowders).

4. Conclusions

The structure and properties of materials on the basis of \( \text{Si}_3\text{N}_4 \) depend on their chemical composition. If the needle-shaped structure prevails in the case of \( \beta\)- \( \text{Si}_3\text{N}_4 \) and \( \beta\)- sialons, than for \( \alpha\)- sialon differing grain forms are possible. This also determines properties of materials: \( \beta\)- \( \text{Si}_3\text{N}_4 \) and \( \beta\)-sialons have mean hardness (17-19 GPa), but high bending strength (700-1100 MPa), whereas \( \alpha\)-sialons have less bending strength (550-700 MPa), but higher hardness (21-22 GPa). The proportion of needle-shaped crystals increases with the rising of sintering temperature with the following increase of bending strength and falling of hardness. Grain size of sialon materials obtained from nanopowders by hot pressing in comparison with materials of similar composition obtained by sintering at 1700 °C is comparatively high (0,5-1,0 \( \mu \)m), but the length of needle-shaped crystals is of 5-10 \( \mu \)m.

References

[1] Yu.G.Gogotsi, J. Mater. Sci., 2000, 29, 2541-2556.
[2] S.A.Suvorov, N.V.Dolgoshev, A.I.Ponikarovsky, A.V.Domakova, K.D.Prohorov, Ogneupory i technitcheskaya keramika, 2006, (3) 2-5 (in russian).
[3] H.Zhao, C.Wood, Y.-B.Cheng, Mat. Sci.Forum, 2000, 325-326, 213-218.
[4] W.T.Young, L.K.L.Falk, H.Lemercier, et.al. J. of Non-Crystalline Solids, 2000, 270, 6-19.
[5] J.Kim, A.Rosenflanz, I.-W.Chen, J. Am. Ceram. Soc., 2000, 83 (7) 1819-1821.
[6] N.Zhilinska, I.Zalite, A.Krumina, W.Costin, G.Mozdzen, In Verbundwerkstoffe, 14. Symposium Verbundwerkstoffe (Degischer, H.P., DGM). WILEY-VCH, 2003, 399-404.
[7] I. Zalite, J. Grabis, Mat.Sci.Forum, 2007, 555, 267-272.
[8] I. Zalite, N. Zhilinska, G. Kladler, Powder Metallurgy Progress, 2006, 6 (2), 88-93.