Properties of Powders and Sintered Bodies of Sialon, Si_3N_4 and SiC Prepared from Korean Raw Materials

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I. INTRODUCTION

The increased demand for Si_3N_4-based materials and SiC as high temperature structural ceramics has focused attention on the development of powder preparation and sintering techniques for non-oxide materials.

The primary object of this work is to produce Si_3N_4, Sialon and SiC powder and sintered ceramics from Korean natural resources and to estimate their applicability to practical uses.

2. RAW MATERIALS

Korean natural resources, i.e., Wando pyrohphyllite, Kimcheon quartzite and Jechon quartzite were selected as starting raw materials to produce β-Sialon, Si_3N_4 and SiC powers.

The chemical compositions of the starting raw materials are given in Table 1. Powders of all starting raw materials were passed through 325 mesh Tyler standard screen before subjected to reaction.

Table 1. Chemical Composition of Raw Materials

Raw Materials	Component (wt%)					
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Ig. Loss
Wando Pyrophyllite	80.32	15.82	0.13	Tr.	0.64	3.06
Kimcheon Quartzite	99.7	0.1	0.04	Tr.	0.01	0.1
Jecheon Quartzite	94.30	2.80	0.70	0.93	Tr.	0.63

Table 2. Sintering Conditions

Sample	Composition (wt%)	Condition
A	Synthesized Sialon	Pressureless Sintering at 1750°C for 90 min
B	Synthesized Sialon	Hot Pressing at 1750°C for 60 min
C	Sialon: MgO = 100: 3	

Table 3. Comparison of properties measured in this work with the values of references

Material	Density (g/cm^3)	R. Temp. M.O.R. (Kpsi)	K_{IC} ($MN/m^{3/2}$)	Hardness (GN/m^2)	Reference
W. Pyrophyllite	2.94	32.9	2.9	12.1	Present work (Sample A)
W. Pyrophyllite	3.08	48.6	4.6	15.3	Present work (Sample C)
Si_3N_4, AlN, Al_2O_3	3.00	51.1	2.21	–	5
Si_3N_4, AlN, Al_2O_3 (SYALON)	3.2	120	5	17.6	6
Si_3N_4 (RBSN)	2.4-2.8	28.4-42	3.6	8.8-10.7	7

3. EXPERIMENTAL PROCEDURE

3.1 β-Sialon from Wando Pyrophyllite

Graphite Powders were excessively added to Wando pyrophyllite powders to produce β-Sialon powders by carbothermal reduction and nitridation at 1350°C in $N_2(80)$ - H_2 (20) atmosphere.[1]

The β-Sialon powers obtained from Wando pyrophyllite were sintered at 1750°C by pressureless sintering for 90 min and by hot pressing with or without MgO as a sintering aid.[2]

3.2 Silicon Nitride from Kimcheon Quartizite

Carbon black powders were excessively added to Kimcheon quartzite to produce silicon nitride powders at 1400°C in hydrogen atmosphere.

The obtained Si_3N_4 powders were acid treated and sintered at 1780°C in nitrogen atmosphere using Y_2O_3 as a sintering aid.[2]

3.3 SiC powder from Jecheon Quartzite

Carbon black powders were excessively added to Jecheon quartzite powders to produce SiC powders at 1400°C in hydrogen atmosphere.

The obtained SiC powders were acid treated before hot pressing at 1900°C.

4. RESULTS AND DISCUSSION

4.1 β-Sialon from Wando Pyrophyllite

The obtained β-Sialon powders were observed to be acicular shaped and very fine (mostly a few μm).

The X-ray diffraction pattern of the obtained β-Sialon powders shows that a little amount of AlN and α-Si_3N_4 is contained as given in Fig. 1.

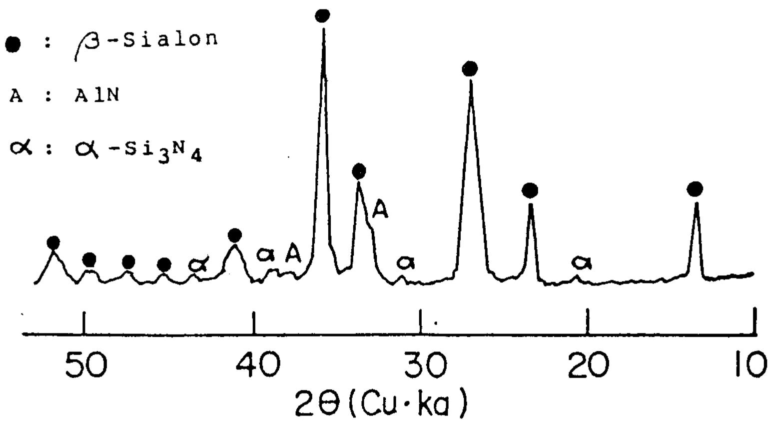

Fig. 1. X-ray diffraction pattrern of the reaction product when W. Pyrophyllite/carbon (SiO_2/C=1/1.2) mixture was heated at 1350°C for 10h in $80N_2$-$20H_2$.

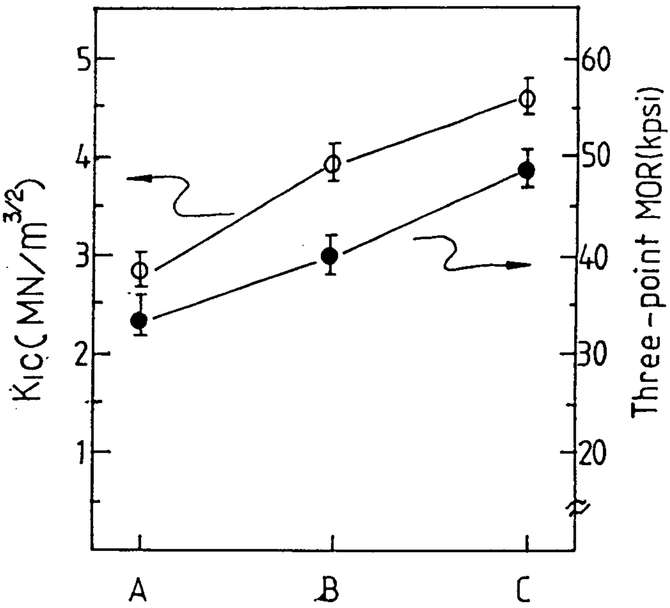

Fig. 2.　K_{IC} and MOR after sintering at 1750°C.

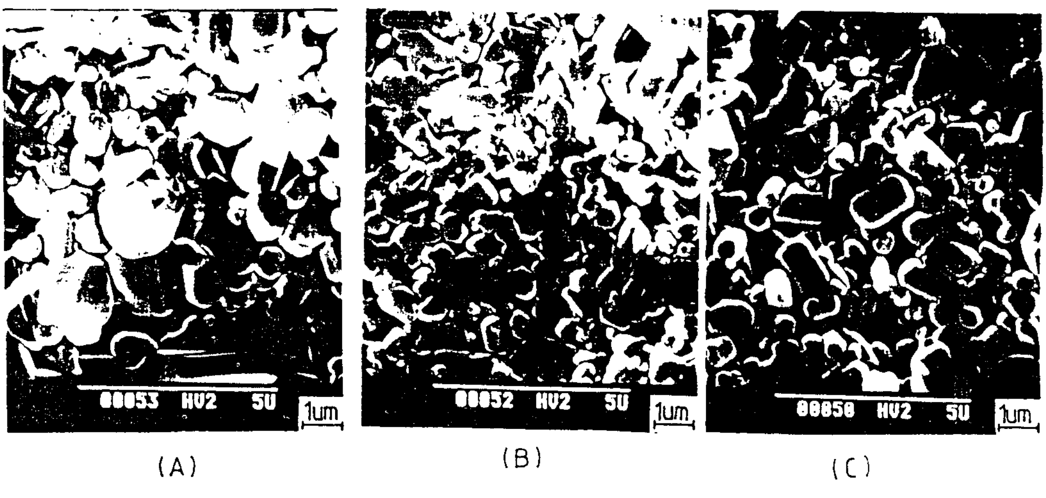

Fig. 3.　SEM photographs of fracture surfaces.

The β-Sialon powders were sintered under the conditions given in Table 2. The densities of sintered bodies were higher than 94% of theoretical values, and the porosities were less than 0.4% for all conditions of sintering.

The X-ray diffraction analysis revealed that all samples had a little amount of X-phase besides β-Sialon, the major phase. AlN and α-Si_3N_4 contained in β-Sialon powers were all dissolved into β-Sialon during sintering at 1750°C. A little amount of X-phase was produced probably because of surface silica.[3]

The values of M.O.R. and fracture toughness(K_{IC}) were measured and given in Fig. 2. The sample hot-pressed with MgO addition represents 48.6 kpsi of M.O.R. and 4.6MN/$m^{3/2}$ of K_{IC}. As can be seen in Fig. 3, sample B, hot-pressed without any additive, has more homogeneously grown grains compared with the pressurelessly sintered sample A. Sample C, hot-pressed with MgO addition, however, has pillar-shaped grains and represents the highest mechanical properties of all.[4-6] The measured properties for β-Sialon ceramics are compared with those reported in references[5-7] as shown in Table 3. The mechanical properties of β-Sialon ceramics obtained from Wando pyrophyllite are comparable to those of foreign commercial products.

4.2 Silicon Nitride from Kimcheon Quartzite

Time-dependence of reaction products was observed. The X-ray intensity of α-Si_3N_4 increased with time, but it decreased again after 9 hours reaction, instead, the intensity of β-Si_3N_4 increased.

X-ray diffraction pattern of reaction product Si_3N_4 obtained after 10 hours reaction at 1400°C showed that α/β ratio was 64/36.

TEM photography for Kimcheon quartzite and synthesized Si_3N_4 powders revealed that the sizes of particles were much reduced from several tens of μm to a few μm by carbothermal reduction and nitridation. The sizes of these synthesized Si_3N_4 powders were comparable to those of the commercial Si_3N_4 fine powders.

4.3 SiliconCarbide from Jecheon Quartzite

Three kinds of samples; as prepared powder, room temperature acid ($HF+HNO_3$) treated powder and hot acid ($HF+HNO_3$) treated powder were analyzed chemically and their surface areas were measured.

After hot acid treating, most of the impurities were eliminated except 1.5 wt% Al_2O_3 which could be used as the sintering aid. The surface area was increased up to 117m^2/g when the powders were hot acid treated, almost five times that of as prepared SiC powder.

5. SUMMARY

Sialon, Si_3N_4 and SiC powders and ceramics were prepared from Korean natural resources. The properties of the powders and sintered ceramics obtained in this work are considered to be comparable to those of foreign articles.

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