



Influence of vermiculite on the formation of porous cordierites

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ABSTRACT

Porous cordierites were prepared from mixtures of talc, kaolinite and vermiculite with alumina or aluminum hydroxide by sintering in an argon atmosphere at 1300 °C for 1 h (porous cordierites of Group I) and 2 h (porous cordierites of Group II). Group I and Group II were different in pore size distribution, quantitative content of mineral phases and cordierite structure parameters. The crystalline phases identified in porous cordierites prepared without vermiculite were cordierite (69–88 vol.%), protoenstatite (25–13 vol.%) and corundum (3–10 vol.%). Vermiculite increased the content of cordierite (84–90 vol.%) and Mg–Al spinel (4–15 vol.%) at the expense of protoenstatite. The calculated unit cell parameters of cordierites were between those of orthogonal and hexagonal cordierites (Group I) and corresponded to disordered orthorhombic rather than hexagonal cordierites (Group II). Group I cordierites had pore size maxima at about ~35 μm or ~20 μm, while Group II had maxima at about 35 μm, 25 μm and 18 μm. Porous cordierites prepared from alumina admixture had slightly lower pore volumes than those prepared from gibbsite. Cordierites prepared from mixtures rich in vermiculite and gibbsite showed the highest porosity (63% and 57%).

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1. Introduction

Ceramic cordierites have low thermal expansion coefficient, excellent high temperature properties, and good surface properties. The most commonly prepared cordierite in the system MgO–Al₂O₃–SiO₂ is often accompanied by spinel, cristobalite, mullite, forsterite, periclase, and corundum (Smart and Glasser, 1976). The conventional methods for the synthesis of cordierite ceramics include solid-state sintering of magnesium, aluminum and silicon oxide corresponding to chemical composition of cordierite (2MgO·2Al₂O₃·5SiO₂). Porous cordierites consisted of α-cordierite and a small amount of spinel and enstatite and were synthesized by sintering of kaolinite and talc (Nakahara et al., 1995; Trumbulovic et al., 2003; Goren et al., 2006a,b) and also Mg-vermiculite (Valášková and Simha Martynková, 2009). Other cordierites were synthesized from the mixtures of clay, talc, alumina, and silica sand (Alves et al., 1998), kaolin, quartz, technical silica, or from talc, kaolin, silica, feldspar (Acimovic et al., 2003), from talc, fly ash, fused silica, and alumina (Kumar et al., 2000). Kaolinite and magnesium hydroxide were sintered to compact cordierites (Kobayshi et al., 2000).

Cordierite forms by exothermic reaction at ~1300 °C (Lamar and Warner, 1954). Mg-cordierites crystallize in two polymorphs. Hexagonal α-cordierite is stable above 1450 °C (Schreyer and Schairer, 1961; Putnis, 1980b). An orthorhombic β-cordierite is stable between 1450 °C and 1460 °C (Meagher and Gibbs, 1977; Cohen et al., 1977). Miyashiro (1957) uses the term “indialite” for samples isostructural with beryl of

hexagonal symmetry and the term “cordierites” for orthorhombic forms. Mg-cordierite when rapidly crystallized from a glass of cordierite stoichiometry is well crystalline, hexagonal, defect-free and homogeneous (Putnis and Bish, 1983). The crystal structure of cordierite was investigated by Rankin and Merwin (1918), Takane and Takeuchi (1936), Byström (1942), Gibbs (1966), Cohen et al. (1977), Meagher and Gibbs (1977), Schwartz et al. (1994). Mg-cordierites have a tetrahedral silicates framework with the simplified structure formula Mg₂Al₄Si₅O₁₈. The Al and Si atoms are distributed over tetrahedral (T) sites: three T1 and six T2 sites per formula unit; Mg atoms are located in slightly flattened octahedra. Many studies reported changes of the tetrahedral Si/Al ordering and transition to the ordered form (Schreyer and Schairer, 1961; Putnis, 1980a,b; Putnis and Bish, 1983; Carpenter et al., 1983; Putnis et al., 1987; Güttler et al., 1989; Schwartz et al., 1994). Hochella et al. (1979), Wallace and Wenk (1980) and Armbruster (1985) studied substitution of structural iron in Fe-cordierites. The presence of smaller size cations causes flattening of the Mg octahedra, the cell parameter *c* becomes smaller and *a* and *b* becomes larger in comparison with Mg-cordierite. Evans et al. (1980) prepared cordierite by different techniques and studied the thermal expansion using high temperature X-ray diffraction. The unit cell volume increased with the amount of Al³⁺ substituted for Si⁴⁺. The thermal expansion of the unit cell of hexagonal cordierite was investigated by Predecki et al. (1987) using neutron powder diffraction. The decrease of the *c* axis resulted from the distortion of the T2 tetrahedra and the coupled distortion of the edge shearing T1 tetrahedra and M octahedra.

Thermal decomposition above 700 °C of vermiculite results in removal of interlayer water and formation of a highly porous material. Okada et al. (2008) observed that mixing allophane with vermiculite

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Table 1
Composition of the mixtures for cordierite ceramics.

Samples	Mineral content, (mass %)			
	Talc	Kaolinite	Vermiculite	Alumina/Gibbsite
ZA/ZG	40	47	–	13
ZVA/ZVG	30	45	12	13
XVA/XVG	30	20	30	20

could control various pore sizes, providing a simple way for enhancing the water-retention properties.

Vermiculite was sporadically used for cordierite synthesis. The subject of our study is the preparation of porous cordierites from vermiculite, their morphology, porosity characteristics, quantitative content of mineral phases and structure parameters.

2. Materials and methods

2.1. Specimens and sintering

Kaolinite and vermiculite from Czech Republic and talc from Egypt were ground separately in a planetary ball mill for 20 min, and then sieved <40 μm . Powders Al_2O_3 (A, alumina) and $\text{Al}(\text{OH})_3$ (G, gibbsite) were purchased from Sigma-Aldrich, Co. The elemental analysis was obtained by X-ray fluorescence spectroscopy (SPECTRO XEPOS new-energy dispersive X-ray fluorescence spectrometer) and atom absorption spectrometry (UNICAM 989 QZ). The structural formulae were approximately $\text{Si}_2\text{Al}_2\text{O}_5(\text{OH})_4$ for kaolinite, $\text{Mg}_3\text{Si}_4\text{O}_{10}(\text{OH})_2$ for talc and $(\text{Si}_{3.13}\text{Al}_{0.86}\text{Ti}_{0.02})^{\text{IV}}(\text{Mg}_{2.53}\text{Fe}_{0.45}^{3+}\text{Al}_{0.02})^{\text{VI}}\text{O}_{10}(\text{OH})_2(\text{Mg}_{0.19}\text{K}_{0.01}\text{Ca}_{0.02})$ for vermiculite.

Mixtures were prepared in the stoichiometric ratio close to cordierite ($2\text{MgO}\cdot 2\text{Al}_2\text{O}_3\cdot 5\text{SiO}_2$) (Table 1). Kaolinite and talc with alumina (sample named ZA) or aluminum hydroxide (sample named

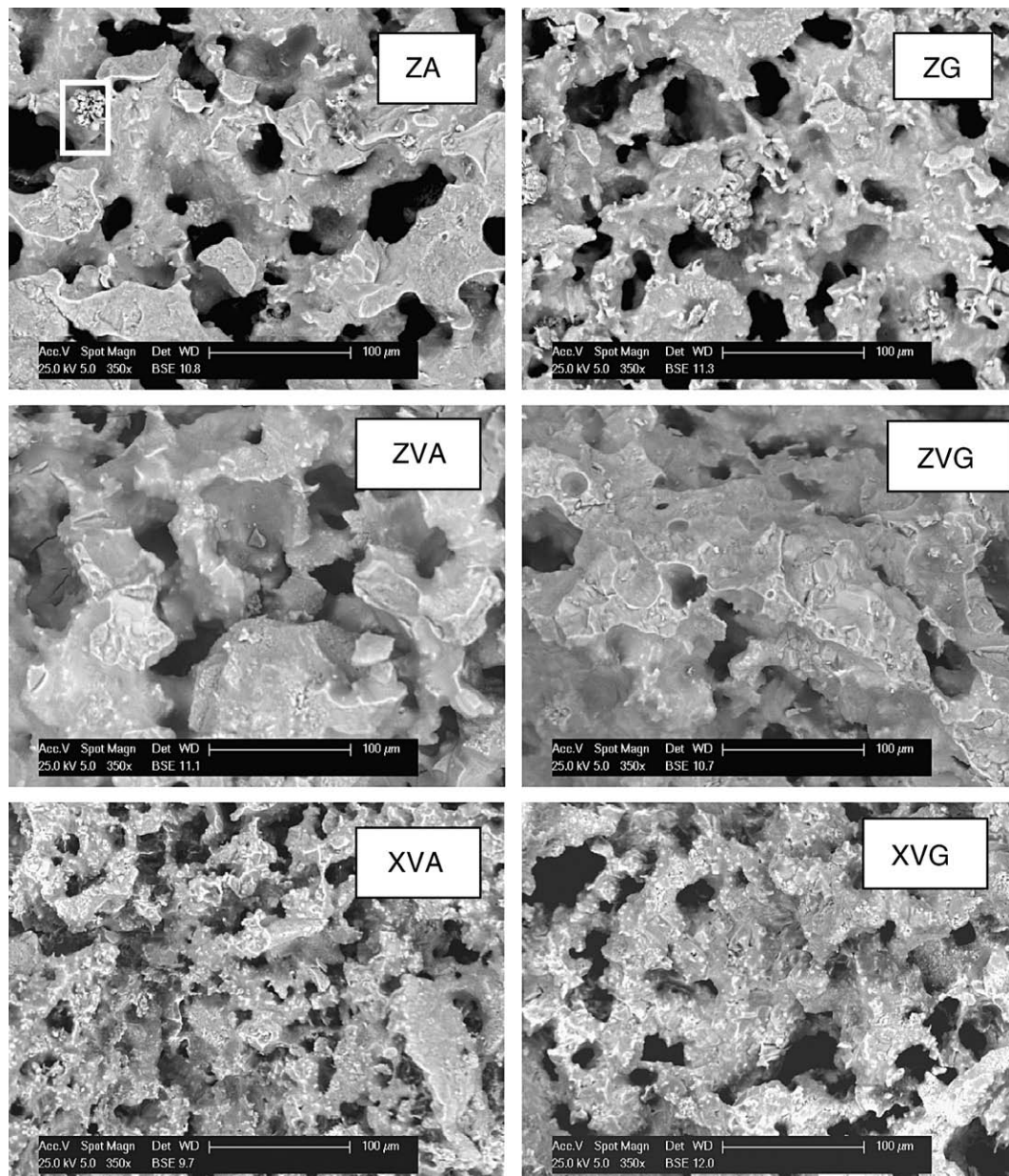


Fig. 1. Scanning electron micrographs of cordierite ceramics. Corundum in ZA is indicated.

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