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Solar sintering of cordierite-based ceramics at low temperatures

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Abstract

Solar furnaces allow materials processing at much higher heating and/or cooling rates compared with those in the conventional industrial and laboratory processes using electric furnaces. During the course of our recent works using solar furnace, we demonstrated usability of solar furnace for sintering-consolidation of oxide ceramics (alumina) and non-oxide ceramics (WC with Co additive) as well as for producing raw material powders including carbide and carbonitride of transition metals and silicon. Being encouraged by these earlier solar-consolidation experiment results obtained for ceramics with relatively simple composition, we decided to continue this line of solar-sintering experiments for other industrial ceramics with more complicated composition. In this work, two types of commercial ceramic powder mixtures, BL7 and RP7, supplied by Rauschert Portuguesa Lda, were subjected to sintering under concentrated solar beam at 950 °C for 20 min. No evidence of formation of cordierite ($2MgO \cdot 2Al_2O_3 \cdot 5SiO_2$) phase was detected for the present solar sintered specimens whereas evidence of cordierite phase formation was detected for the RP7 powders heated to 950 °C in a laboratory electric furnace. As such, the present results implied that, for the preparation of consolidated cordierite using solar furnace, control over the heating rate is of critical importance as well as selection of the starting materials and setting of the processing temperature.

Keywords: Al–Mg–Si–O system; Cordierite; Indialite; Sintering; Solar furnace

1. Introduction

In recent series of works using solar furnace (SF) at PSA (Plataforma Solar de Almería) in southern Spain (Amaral et al., 2000; Cruz Fernandes et al., 1998, 1999, 2002; Guerra Rosa et al., 1999; Martínez et al.,

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1999; Rodríguez et al., 2001; Shohoji et al., 1999, 2000), we demonstrated that concentrated solar radiation can be used as a sustainable ecological heat source for production of carbides (in Ar gas environment) and carbonitrides (in N_2 gas environment) of transition metals from compacted powder mixtures consisting of the metal and carbon (either graphite or amorphous carbon). By the standard 30 min reaction period in Ar gas environment with the maximum heating temperature 1600 °C under solar irradiation, Si and *d*-group transition metals excluding Hf were all fully converted to carbide with high degree of crystallinity. Some combinations of *d*-transition metals and carbon yielded results not readily understandable with reference to available equilibrium phase relationship data suggesting that some photochemical effect besides a thermal effect might be involved in the reaction taking place under certain circumstances of solar heating (Rodríguez et al., 2001).

According to our solar-sintering experiments for oxide ceramics (Cruz Fernandes et al., 2000) and for non-oxide ceramics (Guerra Rosa et al., 2002), the mechanical properties of the ceramic sintered body prepared under solar irradiation were almost comparable to those of the counterpart prepared through conventional industrial sintering processes. Thus, we felt it desirable to further pursue experimental verifications for applicability of SF-sintering for a variety of types of ceramic materials because the quick, cheap, ecological and sustainable nature of solar-sintering would raise productivity of ceramic components with acceptable quality for certain limited applications at reasonable price with minimized environmental load.

In the present study, two commercial ceramic powder mixtures possessing compositions leading to the formation of cordierite ($2MgO \cdot 2Al_2O_3 \cdot 5SiO_2$) phase by heating were sintered at about 950 °C (measured temperature using a type B thermocouple; see Section 3.2.2 for details of the temperature measurement and setting in the present work) in a SF at PSA and the mechanical properties of the sintered disks were evaluated. Phases emerged during the sintering were identified by XRD (X-ray diffraction) analysis. For the sake of comparison, the same two powder mixtures were also sintered at 950 °C in a conventional electric furnace (EF) and subjected to the similar characterization.

There were two main reasons for undertaking the present solar-sintering experiments at such relatively low temperature as 950 °C:

(1) consolidating cordierite at relatively low temperature (<1000 °C) in order to satisfy the industrial demand from the semiconductor sector to fabricate cordierite at temperature lower than the melting points of Cu, Ag or Au;

(2) avoiding undesired melting of the starting materials during heating prior to formation of cordierite ceramics due to inherent difficulty of knowing true temperature in the SF at PSA as explained later in Section 3.2.2.1.

2. Consolidated cordierite-based ceramics

Consolidated cordierite ceramic components appear to have great industrial significance in either dense form (Sumi et al., 1998, 1999) or porous form (Izuhara et al., 2000). Dense sintered bodies of cordierite are used as catalyst carrier for automotive exhaust in form of honeycomb monoliths (Sumi et al., 1998) and also as heat sinks and semiconductor packaging substrates (Sumi et al., 1999) owing to their low dielectric constant (~5 at 1 MHz) and thermal expansion coefficient ($(1–2) \times 10^{-6}\,K^{-1}$ from 25–1000 °C) which closely matches that of silicon. On the other hand, the porous sintered bodies are widely used as high temperature dust collection filters (Izuhara et al., 2000).

For semiconductor applications of cordierite-based ceramics, it is desirable that compositions are tailored to allow sintering to occur at temperatures as low as 1000 °C (below melting temperature of Cu, Ag or Au) making copper co-sintering possible. Unfortunately, it is extremely difficult to produce homogenous and fine cordierite powders that can be sintered without the addition of impurities. Sumi et al. (1999) reported that dense cordierite ceramic bodies could be fabricated by sintering at temperatures lower than 1000 °C from mixtures of kaolinite and magnesium hydroxide together with boron oxide additions. They also reported that mixtures of kaolinite and calcium carbonate without adding the boron oxide additive and fired at a temperature lower than 1000 °C yielded anorthite rather than cordierite (Kobayashi and Kato, 1994).

Natural cordierite is a rare mineral containing significant amounts of iron. Therefore, there is a need to obtain synthetic cordierite from economic raw materials including elementary compounds (e.g. pure oxides, hydroxides and carbonates), double compounds (such as clays, kaolin, talc, steatite) or triple compounds (such as chlorite). Even if silica, magnesia and alumina are added in the right proportions, it is very difficult to sinter pure cordierite without sintering aids because of the very narrow, impurity-sensitive firing range of cordierite (within 25 °C of its incongruent melting point). Additions of K_2O or TiO_2, which increase density from <90% to 98% of theoretical, results in increased dielectric constant, larger thermal expansion and decreased crystallization temperature (Lee and Kriven, 1998). On the other hand, the properties of the final product are dependent upon the extent to which cordierite is devel-