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Liquid phase sintering of SiC with additions of Y_2O_3, Al_2O_3 and SiO_2

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Abstract

Liquid phase sintering of SiC has been studied using different sintering aids with and without the application of pressure during sintering. After Pressureless Sintering the densification behaviour of the SiC-based powder mixtures was observed to be markedly dependent not only on the type and amount of sintering aids used, but also on the crystal structure of the SiC powder. In contrast, hipping leads to high densities in all cases, independently of the chemical composition of the mixture and the type of SiC powder used. When carbon was added to these compositions, fully dense materials were still obtained. However, a pronounced grain growth was observed. Compositions containing additions of SiO_2 and Y_2O_3 did not show any densification after pressureless sintering, due to the high viscosity and extensive volatilisation of the liquids formed at the sintering temperature. On the other hand, fully dense specimens obtained after hiping contained crystalline intergranular phases, which turned to a vitreous structure, as Al_2O_3 was included as a third component.

Keywords: Grain size; Liquid phase sintering; Microstructure-final; SiC

1. Introduction

Liquid phase sintering (LPS) is a well-established route for the consolidation of ceramic materials. Amongst the non-oxide ceramics, silicon nitride (Si_3N_4) containing certain amounts of sintering aids is probably one of the most studied systems.[1–3] In contrast, silicon carbide (SiC) with small additions of boron and carbon has mostly been aimed to be densified in the solid state at very high temperatures, 2000–2200 °C.[4–7] However, the main limitation of the solid-state sintered SiC (SSC) material for some structural applications is its low fracture toughness (3–3.5 MPa $m^{1/2}$), which can be increased, as with Si_3N_4, by the presence of an intergranular phase. Therefore, in the last years the liquid phase sintering of SiC (LPSSC), which can be carried out at lower temperatures (1850–2000 °C), has been investigated.[8–15] Related to the higher fracture toughness of LPSSC (6–7 MPa $m^{1/2}$) the strength can be twice as high.[8,15] On the other hand, the oxidation resistance of LPSSC is inferior in comparison to SSC, mainly due to the presence of the intergranular phase.[16]

Besides, the effect of Hiping up to 200 MPa, under N_2 atmospheres at various temperatures, has also been studied in terms of both β to α transformation and the densification behaviour of these ceramic materials.[17] By analogy with Si_3N_4 it would be expected that the same sintering aids (metallic oxides) could be used for the production of fully dense SiC-based ceramics. Nonetheless, the reduced solubility of SiC in those liquids and the change in crystal structure between its β and α forms, introduce some important differences.

In this work the liquid phase sintering of SiC has been studied using different sintering oxide additives, both by Pressureless Sintering (PS) and by Hot Isostatic Pressing (HIP). The influence of the type and amount of sintering aids, and more specifically the carbon addition, on the densification and the resulting microstructure has been analysed.

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2. Experimental procedure

Commercial powders of β–SiC (Superior Graphite HSC059), β–SiC + 10% α seeds (Lonza QD1) and α-SiC (Lonza UFB10) have been used in this work. Several powder mixtures containing Y_2O_3, Al_2O_3, SiO_2 and C as sintering aids were prepared by wet ball milling during 8 h. The composition and nomenclature of the different powder mixtures are detailed in Table 1.

Powder samples taken from these mixtures were uniaxially pressed at 100–200 MPa to obtain green compacts with green densities between 55 and 65%TD. Sintering was carried out, using two different sintering routes:

1. Pressureless Sintering (PS) in a furnace with graphite elements in closed graphite crucibles in a temperature range between 1700 and 2000 °C during 30 min.
2. Hot Isostatic Pressing (HIP) of glass encapsulated green compacts was carried out between 1700 and 1950 °C, at 100 MPa, during 1 h in an ASEA Q1H-6 HIP equipment.

After sintering, the samples were polished up to 1 μm finish and etched using a boiling Murakami solution (15g KOH + 15g $K_3Fe(CN)_6$ + 60 ml H_2O) during 5 min. The as-sintered microstructures were characterised by SEM, TEM and XRD in order to identify the intergranular phases formed and to determine their chemical composition. For a selection of samples, the final grain size after sintering was measured using a Quantimet image analyser.

Table 1
Nomenclature and chemical composition (wt.%) of samples used

Label	Base powder	C	Y_2O_3	Al_2O_3	SiO_2
β460	β	–	4	6	–
β640		–	6	4	–
β605		–	6	–	5
β625		–	6	2	5
α460	α	–	4	6	–
αC460		4	4	6	–
αC605		3	6	–	5
βα460	β + 10%α	–	4	6	–
βαC460		4	4	6	–
βα230		–	2	3	–
βαC230		4	2	3	–
βα640		–	6	4	–
βαC640		4	6	4	–
βα320		–	3	2	–
βαC320		4	3	2	–
βα605		–	6	–	5
βα625		–	6	2	5

3. Results

3.1. Densification behaviour

Samples of SiC with compositions of $Y_2O_3 + SiO_2$ and $Y_2O_3 + SiO_2 + Al_2O_3$ do not show any significant densification during pressureless sintering within the experimental temperature range used. In this case it is believed that the chemical reaction between C (either, in gaseous form coming from the graphite crucible, or as originally added to the powder mixture) with SiO_2 results in the formation of Y_2O_3-rich liquids at the sintering temperatures with high viscosities and poor wetting characteristics, therefore preventing densification.[18,19] On the other hand, controlled additions of Y_2O_3 and Al_2O_3 to the original SiC powders, allow densification when sintering between 1750 and 1800 °C, depending on the SiC powder used (Fig. 1).

As can be appreciated in Fig. 1 (Fig. 1a, b, c: data for β–SiC, α-SiC and β + α–SiC powders respectively), the densification behaviour of the as-received SiC powders depends on their crystal structure. In this way, for β–SiC powder mixtures a maximum density of 95%TD for β460 (the composition with highest concentration of alumina), is obtained at 1750 °C. In contrast, for pure α or β + α powders the densities obtained are higher than 95%TD for several compositions, when sintering is carried out between 1800 and 1850 °C. It is noteworthy, however, that powder mixtures containing a high concentration of Al_2O_3 (6 wt.%) reach high densities (>90%TD) independently of, both, the addition of C and the original SiC powder used.

It may also be seen, as shown by Fig. 2, that hipping encapsulated green compacts makes it possible to attain nearly full density in all cases, despite the chemical composition and the type of SiC powder used. Nevertheless, it must be emphasised that fully dense LPS SiC-ceramics are preferably obtained, particularly when relatively low temperatures are used, for those samples containing the larger amounts of liquid.

3.2. Microstructure

3.2.1. Pressureless sintering

As mentioned before only compositions from the SiC–Y_2O_3–Al_2O_3 system can be densified by PS, therefore only those microstructures are presented.

Within the temperature range of 1700–1800 °C $3Y_2O_3$ $5Al_2O_3$, yttrium–aluminium–garnet (YAG), is formed as an intergranular phase for all the experimental compositions and independently of the SiC powder used. However, the amount of YAG formed decreases as the sintering temperature was increased. In general more YAG is formed as the Y_2O_3 /Al_2O_3 ratio approaches 0.84 (e.g. 230 and 460 samples). When sintering is carried out at temperatures above 1800 °C, $AlY_3C_{0.5}$ and

$Al_2Y_4O_9$ are formed in increasing amounts as the sintering temperature is also increased.

As observed in Fig. 3, the evolution of the crystalline structure of SiC with sintering temperature, analysed by XRD, depends on the initial structure of the SiC powder used.

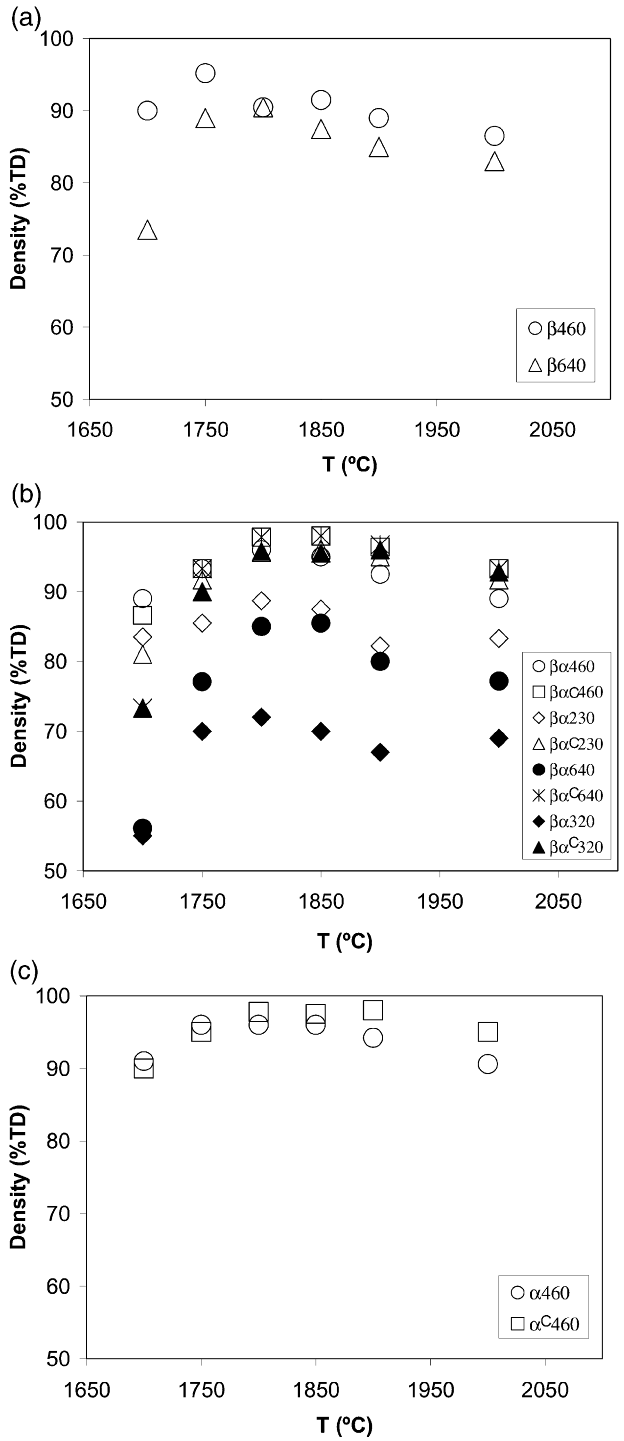

Fig. 1. Dependence of final density as a function of temperature after pressureless sintering of: (a) β–SiC, (b) β+α-SiC and (c) α-SiC powders with $Y_2O_3+Al_2O_3$ additions.

3.2.1.1. Pure β-SiC powders. The β-SiC polytype (3C) shows high stability up to 1750 °C (Fig. 3a). At this sintering temperature the β → α transformation starts to take place, leading to the formation of the 4H α-SiC polytype as the main product. As observed, this transformation is enhanced with temperature, since at 1900 °C nearly all 3C polytype has been transformed, while full transformation is obtained at 2000 °C. Once the β → α transformation takes place, the predominant

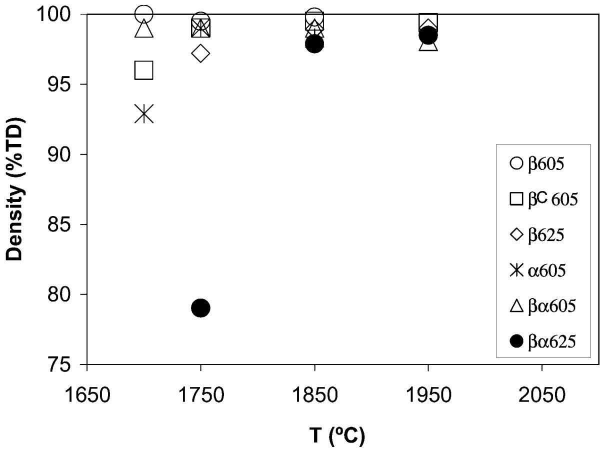

Fig. 2. Final density after HIP for SiC based ceramics with additions of $Y_2O_3+SiO_2$.

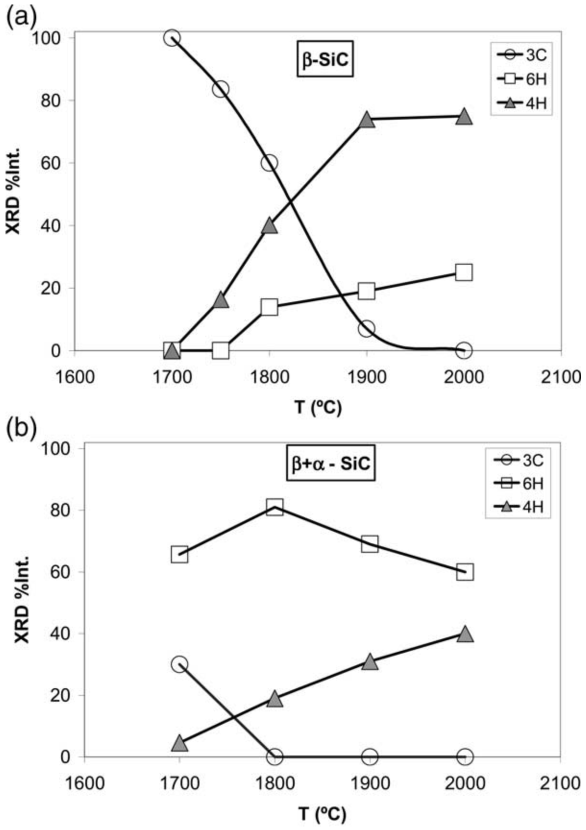

Fig. 3. SiC polytype evolution with temperature for (a) β460 and (b) βα460 compositions.

α polytype observed is 4H (with some amount of 6H polytype) at all temperatures.

3.2.1.2. β + α-SiC powders. In this case, due to the presence of 6H seeds in the original powders, the β → α (mainly 3C → 6H) transformation starts at lower sintering temperatures (< 1700 °C), as compared to pure β–SiC powder mixtures, being completed at 1800 °C. It is worth emphasising that as shown in Fig. 3b from this temperature the 6H → 4H transformation takes place, resulting in a final microstructure consisting of a mixture of the 4H and 6H polytypes, in which the relationship 4H/6H increases with temperature.

With respect to the grain size and morphology, as may be expected, both, the original SiC powder used and the sintering temperature influence the final microstructure of the specimens in the as-sintered condition. This is illustrated in Figs. 4–6, which clearly show the morphology of the resulting grains and their grain sizes. As observed in these figures, sintering of pure α-SiC powder shows a distinct behaviour since the grains remain nearly equiaxed independently of the sintering temperature. In contrast, as appreciated in Figs. 4 and 5 the final microstructure obtained with β or β + α-SiC powders results, as a consequence of the β → α transformation, in nearly all cases, in the development of elongated grains. It may also be seen, by comparing all these micrographs that β and β + α powders are prone to grain growth, whereas specimens sintered from pure α powders show much less grain growth as a function of temperature. This may be appreciated in graphic form in Fig. 7 by comparing the evolution of the grain size (equivalent diameter) with temperature for pure α samples (α460) to those of β + α (βα460). Besides, as carbon is intentionally added to the powder mixture (c.f. βαC460 and βα460) grain growth is encouraged.

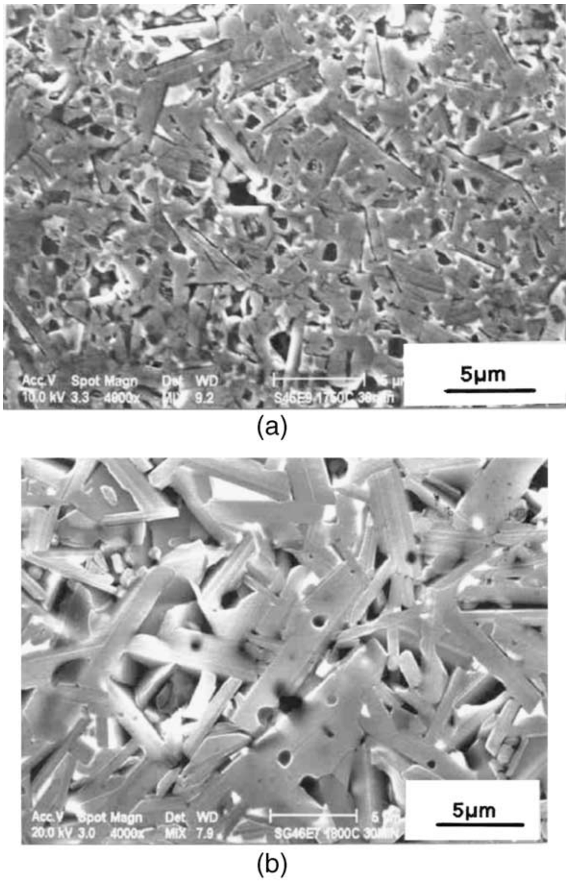

Fig. 4. SEM micrographs showing the morphology of SiC grains after pressureless sintering of an originally β-SiC based powder mixtures (β460) at (a) 1750 °C and (b) 1800 °C.

3.2.2. HIP

As mentioned before, pressureless sintering of specimens containing additions of Y_2O_3 and SiO_2 essentially show no densification at all because of the evaporation of SiO_2 at the sintering temperature. In contrast, the use of hipping, applying a pressure of 100 MPa, allowed full density to be achieved in these specimens. In this case, the solidification of the liquid phase formed at the hipping temperature led to the formation of crystalline intergranular phases at all temperatures: however, the chemical compositions of the intergranular phases formed in the specimens with and without carbon additions were different, since for high carbon contents the resulting intergranular phase was observed to be richer in Yttrium (85% at Y/15% at Si).

On the other hand, it must be emphasised that the crystallinity of the intergranular phases obtained in these specimens was disrupted as Al_2O_3 was added as a third addition to the powder mixture. For these specimens in all cases a glassy intergranular phase was observed, as determined by different authors,[20–22] and its composition, determined in a semi-quantitative manner by EDS, may be written as 13.8 at% Y, 15.7 at% Si, 7.5 at% Al, 63.1 at% O. It may also be mentioned that the $SiC + Y_2O_3–Al_2O_3$ compositions (without SiO_2) produce specimens with YAG as the intergranular phase, the same as by PS, formed in all the experimental temperature ranges used. Furthermore, unless the original SiC powder contains some α seeds, no transformation is observed under any hipping conditions.

4. Discussion

As mentioned in the results section, Fig. 1a–c. show a summary of the results obtained during densification of SiC by the incorporation of various liquids formed within the $Y_2O_3–Al_2O_3$ system. It may also be pointed out that the effect that extra C additions have on the densification behaviour of pure α-SiC and β + α SiC powders has also been considered. By inspection of the density-temperature plot in Fig. 1b, it may, first of all, be noticed that the compositions selected nearly in the stoichiometric ratio Y_2O_3/Al_2O_3 to form YAG (βα640 and βα320 samples) do not produce an important densification within the experimental temperature range used (1700–2000 °C). At temperatures lower than the formation of the liquid (the lowest eutectic temperature for the system Y_2O_3/Al_2O_3 according to the phase diagram and different authors [12, 13] is 1760 °C for 60 wt.% Al_2O_3, Fig. 8[23]) enhanced diffusion does not take place. On the other hand, although in the medium temperature range (1800–1850 °C) a maximum in the densification curve is observed, due to a higher activation of the mass

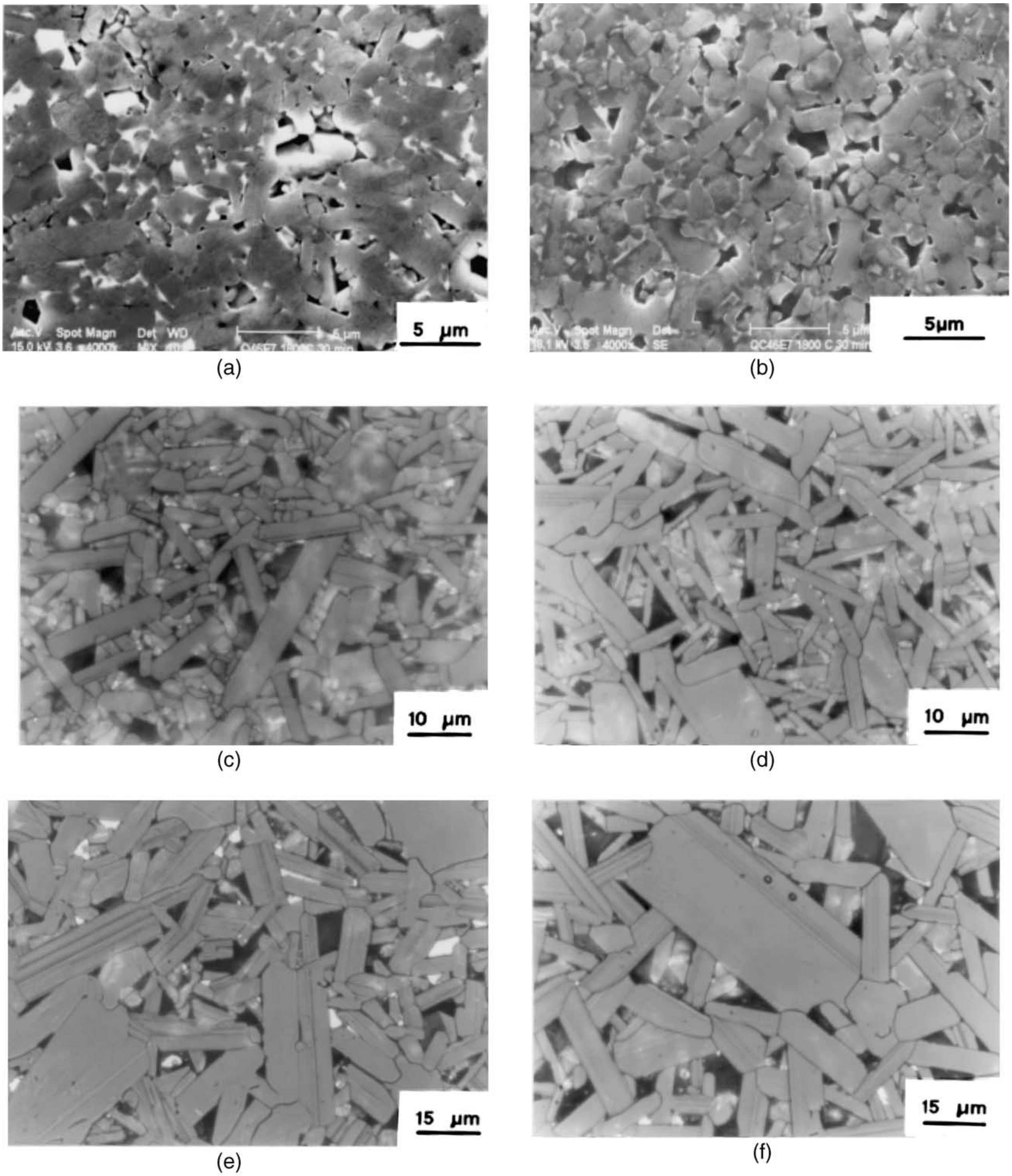

Fig. 5. SEM and Optical micrographs showing the morphology of the hexagonal SiC grains after PS of (β + α)-SiC based powder mixtures, βα460 (a, c, e) and βαC460 (b, d, f) at 1800 °C (a, b), 1900 °C (c, d) and 2000 °C (e, f).

transport due to a possible liquid formation, the density attained is not higher than 85% TD for the ceramic with larger amounts of additions (βα640) and not higher than 70% TD for the specimen labelled as βα320. At higher temperatures it is also worth noting that SiC grain growth, mainly associated to the β→α transformation and more specifically to the 6H→4H polytype transformation, is now encouraged and acts as a competing phenomenon to densification, therefore causing less densification, along with higher weight losses as temperature is increased.

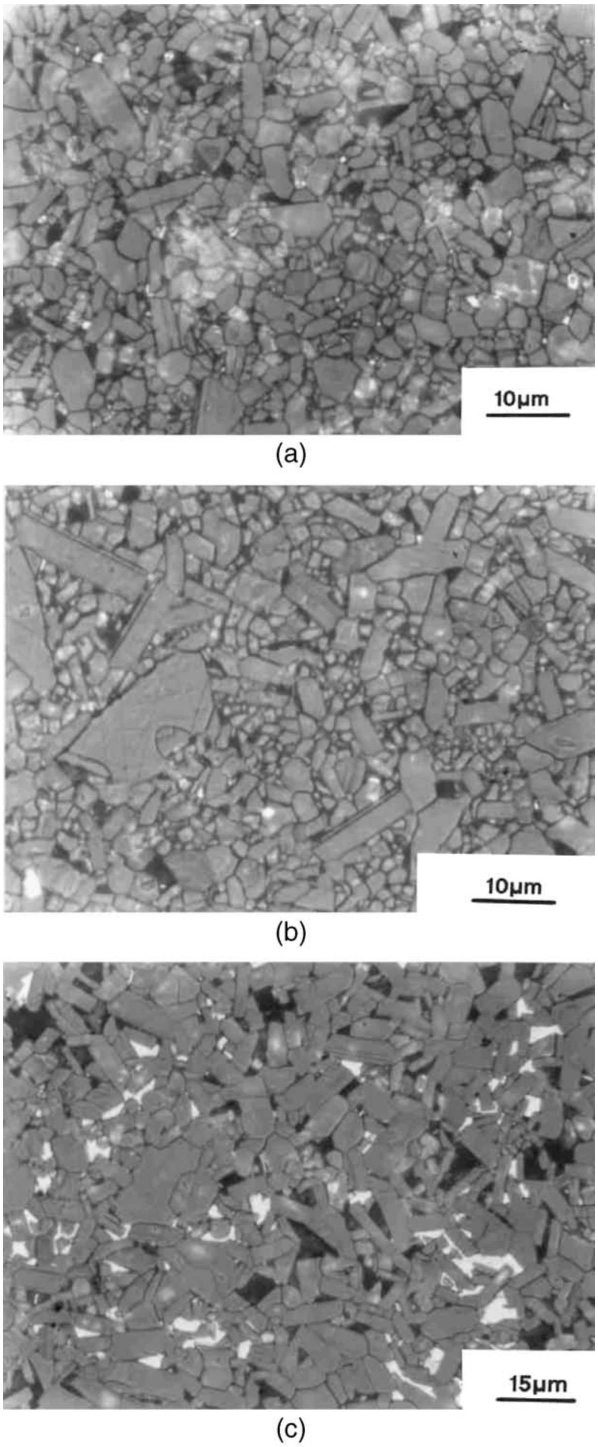

Fig. 6. Optical micrographs showing the morphology of the hexagonal SiC grains after PS of the α-SiC based powder mixture α460, at (a) 1800 °C, (b) 1900 °C and (c) 2000 °C.

In the same figure it is apparent that the specimens containing larger amounts of Al_2O_3 (βα460 and βα230), which have a composition close to the composition of the eutectic liquid (Al_2O_3/Y_2O_3: 60/40), show, as may be expected from the Y_2O_3–Al_2O_3 phase diagram (Fig. 8), a higher final density than those having Y_2O_3–Al_2O_3 in the YAG stoichiometric ratio, particularly in the mid temperature range (1775-1850 °C), above the eutectic temperature. Nonetheless it may also be pointed out that for all compositions the use of too high sintering

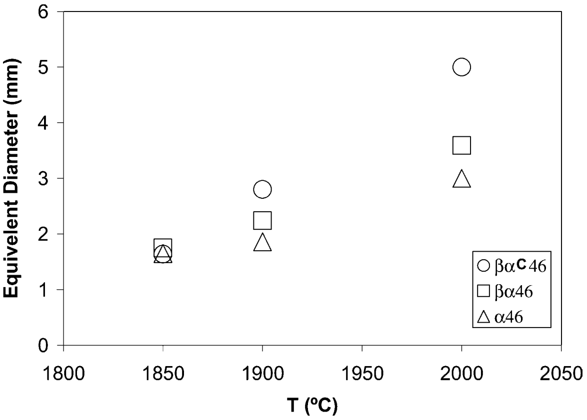

Fig. 7. Mean grain size, expressed as equivalent diameter, of pressureless sintered SiC with $Y_2O_3+Al_2O_3$ additions.

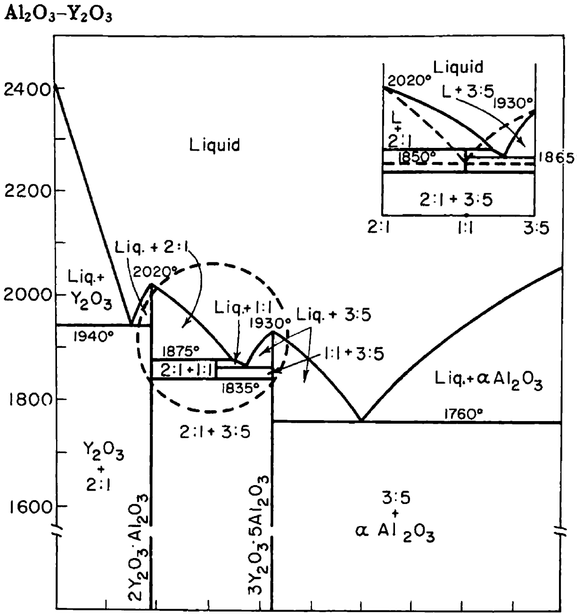

Fig. 8. Y_2O_3/Al_2O_3 phase diagram,[23] (in wt.%).

temperatures again encourages SiC grain growth so hindering densification.

Additionally, it can also be seen that adding C to these basic compositions always produces a beneficial effect on densification, probably due to the formation of liquids of lower viscosity as reported by Misra,[24] who studied the densification of SiC with additions of Al_2O_3 and C. It is important to point out that in the present work the enhancement in densification due to the addition of C to the system $SiC–Y_2O_3–Al_2O_3$ takes place at even lower temperatures than those reported by this author.

It may also be worth mentioning that, as reported elsewhere[8,13,14] important weight losses were observed, ranging from 2 wt.% at 1750 °C to 12 wt.% at 2050 °C, during Pressureless Sintering of these specimens (Fig. 9). Besides, as the amount of Al_2O_3 and C was increased the weight losses observed also increased. These losses may be understood due to the progress of several possible chemical reactions taking place in the experimental temperature range used. In contrast, at the lower temperatures, the SiC/Al_2O_3 interface becomes unstable when carbon activity is high and Al_2O_3 would be converted to Al_4C_3. On the other hand, at higher temperatures, as reported by Van Dijen[8] the reaction:

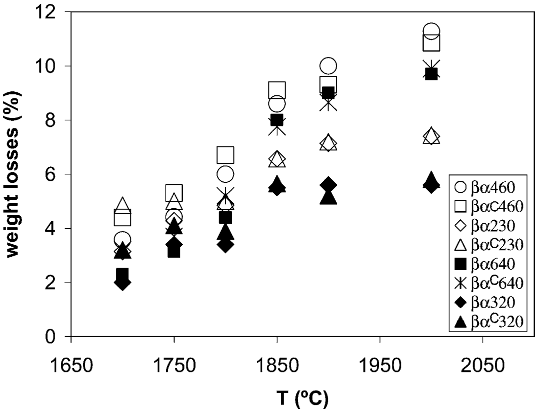

Fig. 9. Weight losses measured for several specimens as a function of sintering temperature.

$$SiC + Al_2O_3 \rightarrow SiO_{(g)} + Al_2O_{(g)} + CO_{(g)}$$

may be taking place causing the volatilisation of gaseous species like SiO, Al_2O and CO. Nevertheless, as shown in Fig. 10, in the present work it was clear that adding Y_2O_3 had an additional effect leading to the formation of Al_2Y_6C, probably as a result of the following reactions:

$$Al_2O_3 + C \rightarrow Al_2OC + O_{2(g)}$$

$$Al_2OC + 2Y_2O_3 \rightarrow Al_2Y_6C + 5/2O_{2(g)}$$

in which C may participate either, from SiC itself or as the extra C added.

It is also interesting to notice that as a result of these reactions the overall composition of these specimens is shifted towards the Y_2O_3 rich side of the Y_2O_3/Al_2O_3 phase diagram, encouraging, as is also apparent in Fig. 10, the formation of $Al_2Y_4O_9$ as an additional phase. This means that, according to the Y_2O_3/Al_2O_3 phase diagram (Fig. 8), the composition of the intergranular phase of βα320 and βα640, with the additions close to the YAG composition, moves to the $2Y_2O_3$ Al_2O_3 – $3Y_2O_3$ $5Al_2O_3$ stability field, in which the minimum temperature for the eutectic liquid is 1865 °C, thus explaining the low densification observed in the mid temperature range (1750–1850 °C) for these materials in comparison to the higher alumina content materials (βα230 and βα460).

Additionally, it should not be overlooked that the extra C added may have a positive effect on densification by the reduction of SiO_2 so promoting wetting of the SiC grains by the liquids formed at the sintering temperatures.

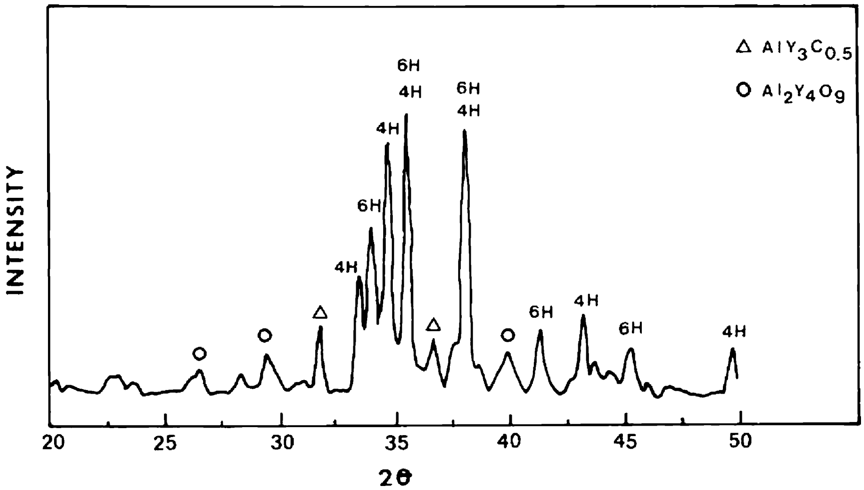

Fig. 10. XRD trace identifying the intergranular phases present in an $Y_2O_3 +$ Al_2O_3 containing SiC specimen after pressureless sintering at 2000 °C.

It is also worth mentioning that by comparing Fig. 1a,b and Figs. 4 and 5, different densification behaviour and microstructure evolution is observed for materials made from β or β + α SiC initial powders. Two main differences can be emphasised between these powders; on the one hand their crystal structure and on the other, the higher free carbon and lower oxygen content of the β–SiC powder from Superior Graphite (1.62 w/o C, 0.9 w/o O), as compared to the β + α SiC powder from Lonza (no free carbon and 1.5 w/o O). The samples made with β–SiC from Superior Graphite can only be densified up to a maximum temperature of 1750 °C. From Figs. 3a and 4b, the polytype transformation 3C → 4H that occurs above this temperature, and may be encouraged by the presence of Al_2O_3 and C, produces an extensive grain growth. This effect counteracts the densification process. In contrast, for the β + α-SiC powder, although the α→ β transformation is completed at 1800 °C (mainly the 3C→6H transformation), the polytype product of the reaction is the 6H, which according to microstructural examinations does not induce significant grain growth at this temperature, thus allowing the densification to progress. Related to this behaviour, the microstructure at 1800 °C consists of equiaxed grains with a much lower final grain size (Fig. 5a) than that of the β–SiC powder mixtures sintered at the same temperature, in which clearly elongated grains are developed (Fig. 4b). Additionally, at higher temperatures the 6H→4H transformation takes place (Fig. 3b) and noticeable grain growth is observed, which is enhanced at these temperatures by the carbon addition, revealed from Figs. 5 and 7.

In contrast, as the specimens are subjected to a HIP treatment, the original β–SiC grains do not show transformation to the hexagonal form and consequently the shape of grains is more equiaxed and grain growth is not as pronounced as before. Under these circumstances, the limited grain growth, together with the application of pressure finally results in fully dense materials.

5. Conclusions

In the present work, it was observed that pressureless sintering of SiC based ceramics in the presence of a liquid phase leads to fully dense materials provided that the liquid formed at the sintering temperature has adequate characteristics, as those formed by sintering aids like alumina and yttria. Additionally, the liquids formed by these sintering aids solidify as crystalline intergranular phases forming the yttrium aluminium garnet (YAG). It must be pointed out that adding SiO_2 to the powder mixture has a negative effect on the densification behaviour of silicon carbide based ceramics.

For all compositions of the powder mixtures and basic SiC powders studied in the present work hipping was revealed to be an effective means to produce fully dense silicon carbide based ceramics by an LPS mechanism. Also, crystalline intergranular phases were observed to be formed in all systems, however, additions of alumina as a third component in the yttria–silica system tend to lower atom mobility thus leading to the formation of glassy phases.

Finally, carbon was revealed as an important sintering aid for this type of ceramics, promoting sintering by reducing the viscosity of the liquid and through the reduction of SiO2, thus improving wetting. In contrast, carbon encourages surface transport mechanisms, thus causing grain growth of the 4H hexagonal polytype at the highest temperatures.

References

1. Iturriza, I., Echeberria, J., Gutiérrez, I. and Castro, F., Densification of silicon nitride ceramics Ander sinter-HIP conditions. *J. Mat. Sci.*, 1990, **25**, 2548–2569.
2. Giachello, A., Martinengo, P. C., Tommasini, G. and Popper, P., *Am. Ceram. Soc. Bull.*, 1980, **59**, 1212.
3. Smith. J. T. and Quackenbush, C. L., A study of sintered Si_3N_4 compositions. In *International Symp. on "Factors in Densification and Sintering of Oxide and Non-Oxide Ceramics"*. Japan Ecl., 1978, pp. 426–442.
4. Prochazka, S., The role of boron and carbon in the sintering of silicon carbide. In *Special Ceramics*, ed. P. Propper. British Ceramic Research Association, Stoke-on-Trent, 1975, pp. 171–182.
5. Greskovich, C. and Rosolowski, J. H., Sintering of covalent solids. *J. Am. Ceram. Soc.*, 1976, **59**, 336–343.
6. Suzuki, H. and Hase, T., Pressureless sintering of SiC ceramics with the addition of B and C. In *High Temperature Ceramic Materials*, ed. H. Suzuki. Nikkan Kougyou Shimbunsha, Tokyo, 1985, pp. 92.
7. Tanaka, H., Sintering of silicon carbide, In *Silicon Carbide Ceramics—1. Fundamental and Solid Reaction*. ed. S. Somiya and Y. Inomata. Elsevier Applied Science, 1991, pp. 213–238.
8. Van Dijen, F. K. and Mayer, E., Liquid phase sintering of silicon carbide. *J. Eur. Ceram. Soc.*, 1996, **16**, 413–420.
9. Liden, E., Carlström, E., Eklund, L., Nyberg, B. and Carlsson, R., Homogeneous distribution of sintering additives in liquid-phase sintered silicon carbide. *J. Am. Ceram. Soc.*, 1995, **78**, 1761–1768.
10. Kostic, E., Sintering of silicon carbide in the presence of oxides additives. *Powder Metall. Int*, 1988, **20**, 28–29.
11. Kostic, E., Nesic, O., and Zec, S., In *Eur. Ceramics. Vol. 1, Processing of Ceramics*. ed. G. De With R.A. and Terpstra, R. Metselaar. Elsevier Applied Science, 1989, p. 1341.
12. Falk, L. K., Microstructural development during liquid phase sintering of silicon carbide ceramics. *J. Eur. Ceram. Soc.*, 1997, **17**, 983–994.
13. She, J. H. and Ueno, K., Effect of additive content on liquid-phase sintering on silicon carbide ceramics. *Mat. Res. Bull.*, 1999, **34**, 1629–1636.
14. Biswas, K., Rixecker, G., Wiedmann, I., Schweizer, M., Upadhyaya, G. and Aldinger, F., Liquid phase sintering and microstructure-property relationships of silicon carbide ceramics with oxynitride additives. *Mat. Chem. Phys.*, 2001, **67**, 180–191.

15. Suzuki, K., Pressureless–sintered silicon carbide with addition of aluminium oxide. In *Silicon Carbide Ceramics, Vol 2*, ed. S. Somiya and Y. Inomata. Elsevier Applied Science, London, 1991, pp. 163–182.
16. Gomez, E., Iturriza, I., Echeberria, J. and Castro, F., Oxidation resistance of SiC ceramics sintered in the solid state or in the presence of a liquid phase. *Scripta Metallurgica et Materialia*, 1995, **33**, 491–496.
17. Gomez, E., Gómez-Acebo, T., Echeberria, J., Iturriza, I. and Castro, F., Nitridation of SiC for the production of SiC–Si_3N_4 nanocomposites. *J. Eur. Ceram. Soc.*, 1994, **14**, 411–418.
18. Mulla, M. A. and Krstic, V. D., Low temperature pressureless sintering of β–SiC with Al_2O_3 and Y_2O_3 additions. *Ceram. Bull.*, 1991, 70.
19. Cutler, R. A. and Jackson, T. B., Liquid phase sintered silicon carbide. In *Proc. 3rd International Symposium on Ceramic Materials and Components for Engines*, ed. V. J. Tennery. American Ceramic Society, Westerwille, OH, 1989, pp. 309–318.
20. Abe, O., Sintering process of Y_2O_3-added Si_3N_4. *J. Mat. Sci.*, 1990, **25**, 3641–3648.
21. Smith, J. T. and Quackembush, *Am. Ceram. Bull*, 1980, **59**, 529.
22. Greskovich, C. and Prochazka, S. J., *J. Am. Ceram. Soc.*, 1977, **60**, 471.
23. Levin, E., Robbins, C., McMurdie. H. M., *Phase Diagrams for Ceramists 1969*. The Am. Ceram. Soc. Pub., Columbus, OH, 1969, p. 96, Fig. 2344.
24. Misra, A. K., Thermochemical analysis of the SiC–Al_2O_3 reaction with reference to liquid phase sintering of SiC. *J. Am. Ceram. Soc.*, 1991, **74**, 345–351.