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B. WILSHIRE,
B. WATKINS,
*Department of Metallurgy and
Materials Technology
University College,
Singleton Park,
Swansea, U K*

Microstructural changes during the argon-sintering of silicon powder compacts

Reaction-bonded silicon nitride (RBSN) is gaining acceptance as an engineering ceramic, and attention is being focused on how processing affects its microstructure [1, 2]. This is because both mechanical properties and oxidation resistance are microstructure-dependent and require optimizing if the material is to be successfully exploited in high temperature engineering.

The fabrication route for RBSN involves the following three main stages: (i) production of the silicon powder compact when particle size, size distribution and the compaction operation are important; (ii) argon-sintering (typically at $\sim 1200^\circ\text{C}$), in which the compact gains sufficient strength to withstand the machining stresses, and (iii) nitriding, during which conversion to the ceramic occurs.

Current Leeds research is concerned with examining the extent to which each stage determines final microstructure. As far as stage (iii) is concerned, the situation is complicated, because the reaction is very sensitive to impurities [3] which are invariably present in significant amounts in the production of commercial material. A firm basis is being established for the control of microstructure during nitriding [4]; the present letter draws attention to some important features of the microstructure developed during argon-sintering, the stage which, as far as the authors are aware, has received little or no attention hitherto.

To investigate the changes in microstructure occurring during this stage, a commercial silicon powder (conventionally ground in a steel ball-mill; analysis in Table I) was isostatically pressed at 200 MN m^{-2} into compacts which were argon-sintered at 1150, 1200 or 1250°C for 1 or 18 h.

TABLE I Analysis of silicon powders

Sample	Composition (Wt %)		
	Ca	Al	Fe
Commercially ground Si (powder 1)	0.20	0.24	0.96
Ceramic ground Si (powder 2)	0.10	1.0	0.31

Chemical analysis at points within the compacts was estimated using the energy dispersive Link System attachment to a Cambridge S600 scanning microscope. Fig. 1a shows the microstructure of a compact before argon-sintering; Fig. 1b is typical of the microstructure developed during argon-sintering.

In a comparison experiment, lump silicon (from the same source as that of Powder 1) was ground in a ceramic mill, to avoid metallic iron contamination (analysis in Table I). The microstructure of a compact of this powder, after sintering in argon at 1250°C for 18 h, is shown in Fig. 1c.

The large voids in Fig. 1b, which were a common feature of these particular samples, were evidently caused by local melting, followed by movement of the melt, due to surface tension forces, into adjacent regions. Each pore had a second phase associated with it, clearly shown up by relief-polishing (Fig. 1d). The composition by weight of this phase was estimated by probe analysis to be Si (51%), Fe (47%) and Al (1.5%). A point by point analysis along a line through the pore confirmed that the iron-rich melt had in fact moved into the surrounding compact (Fig. 2).

The composition of the melt is close to that of the eutectic FeSi_2 [5], having melting point 1212°C . The extent of second phase associated with a pore increased with increasing temperature and its average composition followed the liquidus in the silicon-rich direction. It seems likely that aluminium could account for the melting observed at 1150°C , but this requires confirmation.

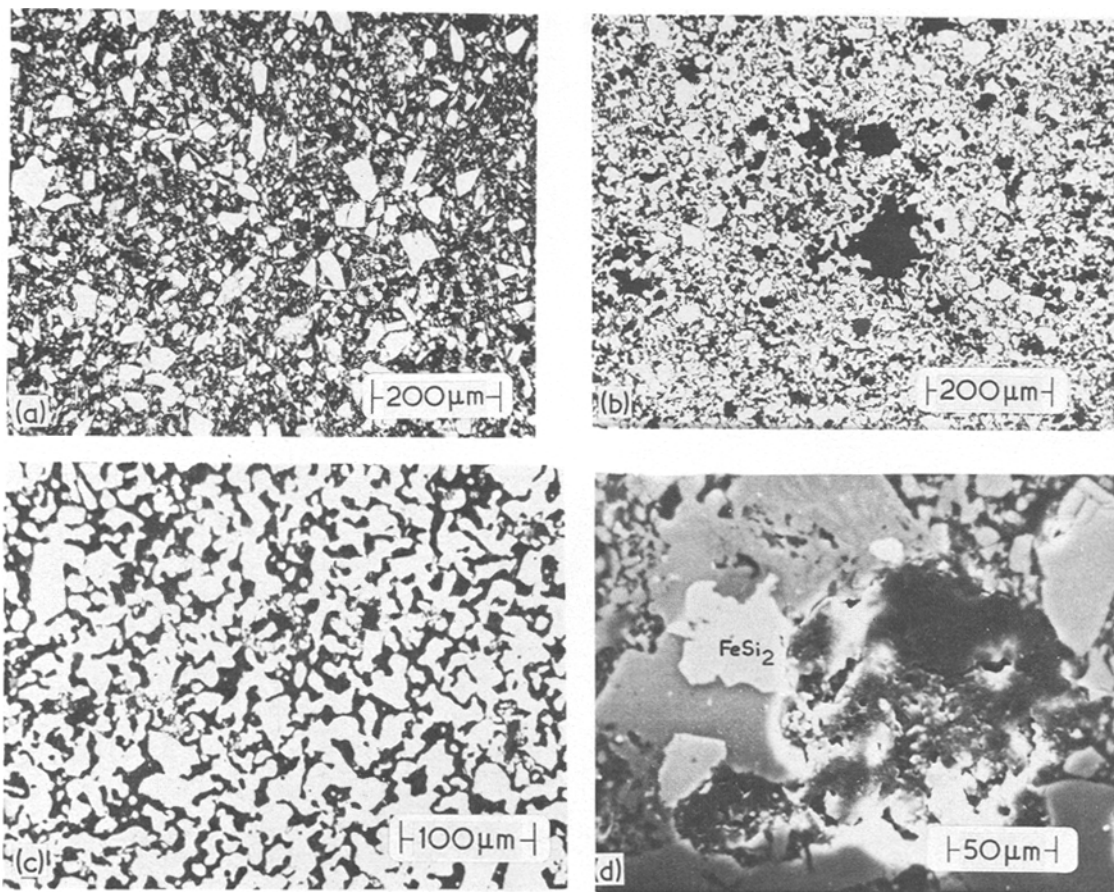


Figure 1 (a) Typical 'green' microstructure prior to argon-sintering (powder 1). (b) Typical microstructure developed during argon-sintering (powder 1); 1 h at 1200° C. (c) Microstructure of argon-sintered compact of specially ground silicon (powder 2); 18 h at 1250° C. (d) Typical defect showing second phase distribution (powder 1); 18 h at 1250° C.

The authors are satisfied that the observations are of general applicability and do occur during the argon-sintering of compacts of the usual commercial powders. It should be borne in mind that

if for any reason (e.g. in the case of injection-moulded or slip cast shapes) the argon-sintering stage is omitted, then the same local melting would occur at the very outset of the nitriding stage.

In summary, the evidence demonstrates that:

(i) particles of iron introduced during the milling of commercial silicon powder cause melting at the argon-sintering (or initial nitriding) stage throughout the compact;

(ii) this local melting produces voids of diameter typically 100 μm;

(iii) the matrix around the pore is modified by penetration of an iron-rich phase, giving rise to inhomogeneities of diameter typically 600 μm;

(iv) these large-scale inhomogeneities, arising prior to the machining and nitriding stages, are of

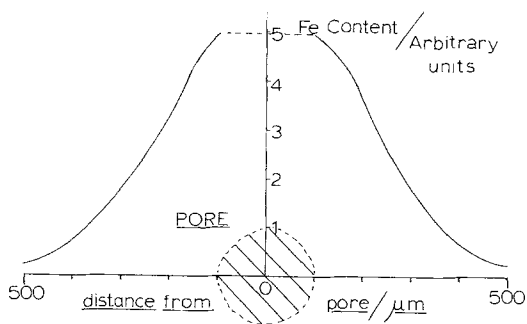


Figure 2 Iron distribution in vicinity of defect (powder 1); 18 h at 1250° C.

a size which warrants their serious consideration as precursors of strength-controlling defects in the ceramic.

It is the opinion of the authors that in any systematic attempt to understand and optimize the mechanical and related properties of RBSN, close attention should be given to these effects and their consequences.

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P. ARUNDALE
A. J. MOULSON
*Department of Ceramics,
Houldsworth School of Applied Science,
The University of Leeds,
Leeds, UK*