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Ceramics International 39 (2013) 6119–6130

CERAMICS INTERNATIONAL

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Carbothermal nitridation process of mechanically milled silica sand using Taguchi's method

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Received 12 November 2012; received in revised form 11 January 2013; accepted 11 January 2013
Available online 29 January 2013

Abstract

This paper reported on the work performed to study the formation of silicon nitride and silicon carbide whiskers using the carbothermal nitridation process. A distinctive aspect of the present study lies in the use of the mechanical milling method to alter the regularity of the crystalline network of the silica sand. In order to optimise the processing parameters for the synthesis of silicon carbide, the concept of Taguchi's Design of Experiments was considered, the analysis being based on Taguchi's signal to noise ratio and variance techniques to obtain optimum combination of process parameters. Important factors influencing the formation of silicon carbide were the duration of the mechanical milling, followed by temperature, time and heating rate.

Keywords: B. Silicon carbide whiskers; Carbothermal nitridation; Taguchi's design of experiments method; Mechanical milling

1. Introduction

Engineering ceramics such as silicon carbide (SiC) and silicon nitride (Si_3N_4) have attractive properties including high strength, stiffness, good resistance to wear and corrosion [1] that renders these materials, especially SiC, as promising candidates for use in thermomechanical and electronic applications [1–4]. In addition, there is a growing demand for these materials to be produced in the whiskers form as reinforcement for composite bodies [5].

Silva and Figueiredo [5] have also studied the Si–C–O–N phase diagram in order to determine the conditions for the formation of each compound from the thermodynamic perspective. According to their findings, there is a limiting temperature above which SiC formation is favoured instead of Si_3N_4. They found that this temperature lies between 1400 °C and 1450 °C, and is strongly influenced by several factors such as the composition of the atmosphere, total pressure, C/SiO_2 molar ratio and pressure impurities [6–9,11]. However, there is some controversy on the subject of which of these reactions accounts for Si_3N_4 whisker formation. It was revealed [5] that silicon monoxide and carbon monoxide acted as intermediates in the formation of both SiC and Si_3N_4, making production a competitive process.

Silicon carbide is an industrially manufactured synthetic product made by the high temperature reaction of high quality silica sand (99.5% SiO_2) with low sulphur petroleum coke. The reaction takes place in an electric resistance furnace and requires large quantities of electricity (23.7 J/kg), where voltage passing through the core produces a temperature of 2400 °C [10,12–18]. As reported by Martin et al. [1], the silicon carbide produced under such conditions possessed large grains and batches which in turn required milling and purification processes to enable further usage. They also reported that the manufacture of silicon carbide whiskers by milling Acheson silicon carbide products had not been successful. Submicron SiC particles can be obtained by several advanced processing techniques such as the chemical vapour deposition process, the sol–gel process and the laser gas phase pyrolysis or

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http://dx.doi.org/10.1016/j.ceramint.2013.01.030

laser evaporation process. Since the most easily scalable process is the carbothermal reduction of silica, efforts have been made to improve silicon carbide particle size control by performing appropriate control to the size of carbon and silica particles inside the reaction mixture [14–22].

The apparent existing problem of the above mentioned technique is the high temperature requirements of about 2400 °C for the reaction to be feasible when using silica sand as the starting precursor [8].

The main objective of this work is therefore to study the effects of four processing factors; temperature, time, heating rate and duration of mechanical milling process for silica sand on the formation of silicon carbide from the carbothermal nitridation process. Experiments were performed based on Taguchi's Design of Experiments method, and the analysis of

Table 1
Processing parameters and their levels for orthogonal experiment.

Column	Process parameter	Level 1	Level 2	Level 3
A (°C)	Temperature	1450	1550	1650
B (min)	Time	60	120	180
C (°C/min)	Heating rate	5	10	20
D (hour)	Duration of mechanical milling process of silica sand	20	60	100

Table 2
Comparisons of factorial design and Taguchi design.

Factors	Level	Total number of experiments	
		Factorial design	Taguchi
2	2	4 (2^2)	4
3	2	8 (2^3)	4
4	2	16 (2^4)	8
7	2	128 (2^7)	8
15	2	32,768 (2^{15})	16
4	3	81 (3^4)	9

Table 3
The L_9 (3^4) *OA* (parameters assigned) with response.

Serial number	Run order	Parameter trial conditions				Responses (raw data)*		Signal-to-noise ratio (dB)
		A(1)	*B*(2)	*C*(3)	*D*(4)	R_1	R_2	
1	3	1	1	1	1	Y_{11}	Y_{12}	*S*/*N*(1)
2	7	1	2	2	2	Y_{21}	Y_{22}	*S*/*N*(2)
3	5	1	3	3	3	Y_{31}	Y_{32}	*S*/*N*(3)
4	1	2	1	2	3	Y_{41}	Y_{42}	*S*/*N*(4)
5	4	2	2	3	1	Y_{51}	Y_{52}	*S*/*N*(5)
6	6	2	3	1	2	Y_{61}	Y_{62}	*S*/*N*(6)
7	9	3	1	3	2	Y_{71}	Y_{72}	*S*/*N*(7)
8	2	3	2	1	3	Y_{81}	Y_{82}	*S*/*N*(8)
9	8	3	3	2	1	Y_{91}	Y_{92}	*S*/*N*(9)

*R_1, and R_2, are responses values for two repetition of each trial. The 1's, 2's and 3's denote the levels 1, 2 and 3 of the parameters. Y_{ij} are measured values of the quality characteristic for the % yield of SiC formation.

Table 4
Experimental results and their *S*/*N* ratios.

Trial no.	Weight of SiC (g)		MSD	*S*/*N*
	R_1	R_2		
1	5.300	5.160	3.658E-02	14.36
2	6.310	6.490	2.443E-02	16.12
3	9.000	8.770	1.267E-02	18.97
4	9.660	9.850	1.051E-02	19.78
5	5.150	4.940	3.934E-02	14.05
6	9.840	9.540	1.066E-02	19.72
7	7.820	7.810	1.637E-02	17.85
8	11.200	11.400	7.833E-03	21.06
9	8.540	8.480	1.381E-02	18.59
Mean				17.83

Table 5
The average effects of factors for each level.

Column	Factors	Average Effects		
		Level 1	Level 2	Level 3
1	Temperature	16.49	17.85	19.17
2	Time	17.34	17.08	19.10
3	Heating Rate	18.38	18.17	16.96
4	Duration of mechanical milling process of silica sand	15.67	17.90	19.94

Table 6
The ANOVA table.

Column	Factors	DOF	SS	Variance	F	%
1	Temperature	2	10.823	5.411	-	22.126
2	Time	2	7.248	3.624	-	14.818
3	Heating rate	2	3.530	1.765	-	7.218
4	Duration of mechanical milling process of silica sand	2	27.313	13.656	-	55.835
	All others/ error	0	–	–		–
	Total	8	48.914			100.00

Table 7
The pooled ANOVA table.

Column	Factors	DOF	SS	Variance	F	%
1	Temperature	2	10.823	5.411	3.065	14.908
2	Time	2	7.248	3.624	2.053	7.600
3	Heating rate	2	3.530	-	-	-
4	Duration of mechanical milling process of silica sand	2	27.313	13.656	7.736	48.620
	All others/ error	2	3.530	1.765		28.871
	Total	8	48.914			100.00

Table 8
Estimate of the optimum condition of design.

Factors	Level description	Level	Contribution
Temperature	1650 °C	3	1.335
Time	180 min	3	1.260
Duration of mechanical milling process of silica sand	100 h	3	2.101
Contributions from all factors (total)			4.697
Current grand average of performance or mean			17.837
Expected results at optimum condition			22.534

variance (ANOVA) technique was employed to ascertain the relevant significance of the processing factors. The present study also aims to assess the formation of silicon nitride and silicon carbide materials at various temperatures derived from the carbothermal nitridation process using as received silica sand and structurally distorted silica sand.

2. Experimental procedure

Silica sand of purity 99.5%, obtained from Johore Silica was screened to less than 32 μm. The carbon powder, which had undergone ball milling treatment for five hours, was obtained from Petronas refinery plant. The C/SiO_2 mixtures in a molar ratio of 6/1 were prepared by mixing in a rotary mixer for 30 min. Two batches of mixture were prepared, i.e. one batch was subjected to mechanical milling (MM) and the other was in 'as received' state (Un-MM). The mechanical treatment of the silica sand was performed in a planetary ball mill of four stations using a stainless steel jar with a ball to powder ratio (BPR) 20:1 by weight and milling speed of 200 rev min^{-1}. The synthesis process was carried out in tube furnaces of 250 cm^3 working volume with mixed gas flow above the layer of mixture. The 1–2 mm thick layer of mixture was placed in boats made of alumina and then put into a heating zone of the furnace. The furnace was purged with mixed gas (5% H_2+95% N_2) for about 15 min before the process. The furnace was heated up to the desired temperature at a chosen rate, then the temperature was kept constant over the desired time. The process was carried out at temperatures ranging from 1350 to 1650 °C, then the samples were furnace cooled. The samples were removed from the furnace, ground in mortar and then analysed according to the prescribed procedure. The particle shape and chemical composition of the product syntheses at various temperatures were determined by scanning electron microscopy (Leo 1510) and X-ray diffraction methods. An elemental analyser, Leco, model TC-300 was used to determine the elemental analysis of C, H and N [5].

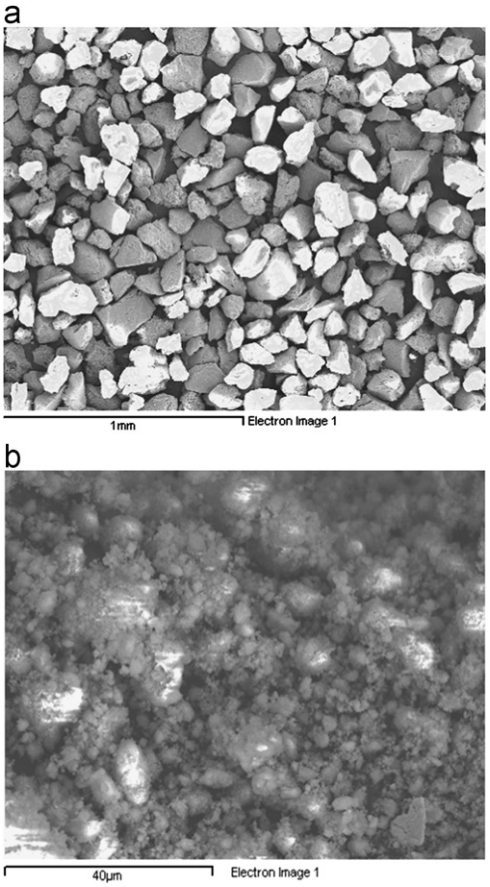

Fig. 1. SEM micrograph of (a) Un-MM silica sand and (b) MM silica sand.

The influence of four factors; temperature, time, heating rate of the process and duration of mechanical milling of silica sand; on the yield of formation of SiC was studied by using the L_9 orthogonal array, with each factor having three levels. The factors involved and their levels are shown in Table 1. If full factorial experimental design was used, it would require 81 trial runs for all possible permutations of these factors [23–30].

In order to minimise the number of experiments needed to produce relevant information for the set of inputs, the Taguchi orthogonal arrays were considered. As such, in this study, with

the use of L_9 orthogonal array, the number of runs was substantially reduced to nine experiments. The comparison between full factorial design and Taguchi design is shown in Table 2 whereas Table 3 presents the L_9 orthogonal array [29]. Since there were four three-level factors, these factors were assigned to all four columns of the L_9 array. In this work, interaction effects between factors were considered insignificant and hence, not examined.

Nine trial runs with certain factor level combinations determined from the array were carried out and duplicated (R_1 refer to the 1st reading and R_2 refer to the 2nd reading). The composition of the products was determined from elemental analysis by weight% of C, N and H in the samples, based on the work of Silva and Figueiredo [5] which used the following assumptions:

- Species considered in the samples: SiC, Si_3N_4, SiO_2 and H_2O (the free carbon was previously burned off).
- All carbon was present as SiC, all nitrogen as Si_3N_4 and all hydrogen as H_2O. Unreacted silica is computed by the weight difference.

It was also assumed that the amount of water calculated was in general less than 0.5 wt% and, therefore, was omitted from the results. The reaction yields were based on silicon and computed using Eqs. (1) and (2) [5]:

$$Total_{yield} = \frac{nSiC + 3nSi_3N_4}{nSiO_2} \times 100 \quad (1)$$

$$SiC_{yield} = \frac{nSiC}{nSiO_2} \times 100 \quad (2)$$

where $nSiC$ is the number of moles of silicon carbide present in the sample and $nSiO_2$ is the number of moles of silica initially present.

3. Results and discussion

3.1. Statistical Optimisation

The results of the nine trial conditions, with two runs per trial condition are given in Table 4. Kumar et al. and Ibrahim et al. [23,24] described three types of quality characteristics

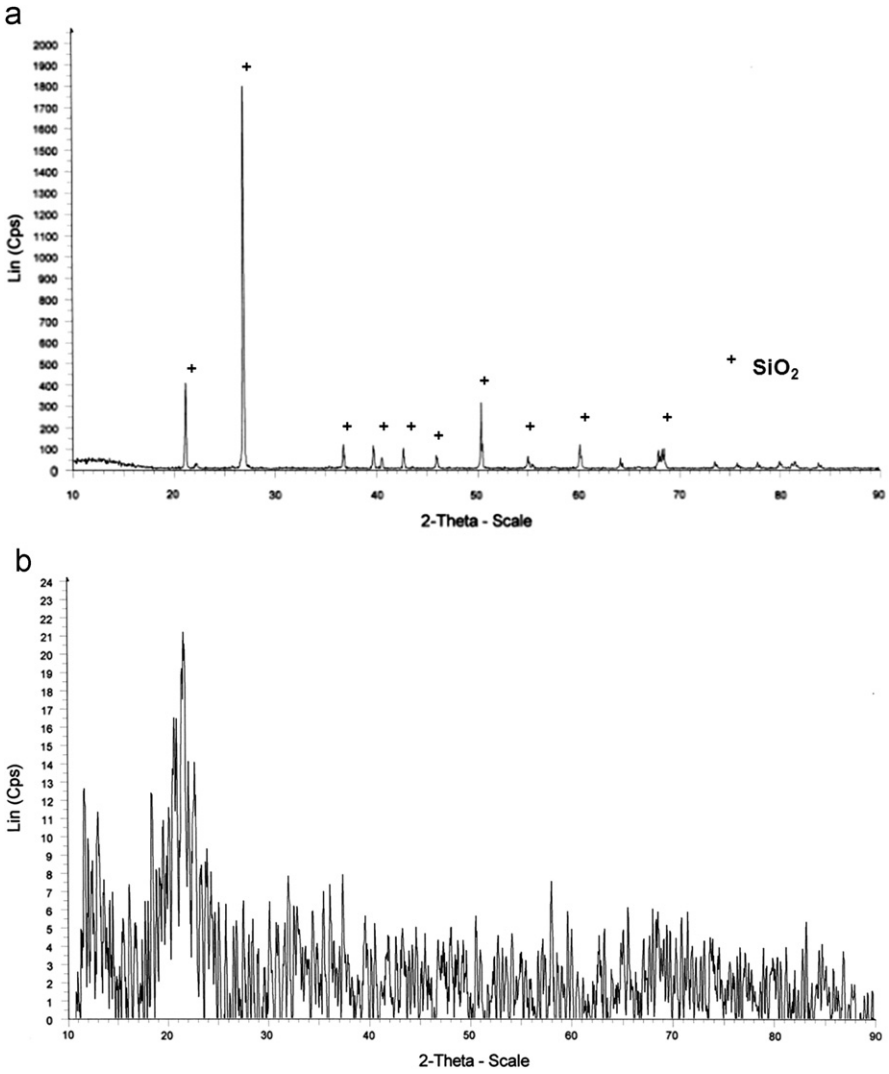

Fig. 2. XRD spectrum of (a) Un-MM silica sand and (b) MM silica sand.

with respect to the target in Taguchi's analysis method, i.e. 'smaller is better', 'nominal is better' and 'bigger is better'. In this study, higher compressive strength was the desired outcome thus, it was categorised in the 'bigger is better' quality characteristic. Hence, in Table 4, the Mean Squared Deviation (MSD) is transformed into signal to noise ratio (S/N). This transformation consolidates the repeated response (raw data) in each trial condition into a single number. Thus, nine S/N ratios were calculated which in turn also reduced the total degree of freedom (DOF) of the experiments. The quality characteristic for the % yield of SiC formation is of the higher-the-better type. So, the signal-to-noise (S/N ratio) for the "higher-the-better" type was used and is shown in Eq. (3):

$$\left(\frac{S}{N}\right)_{HB} = -10\log\left[\frac{1}{n}\sum_{j=1}^{R}\left(\frac{1}{Y_j^2}\right)\right] \tag{3}$$

Table 5 shows that there is a gradual increase in the average effects starting from level 1 to level 3 for factors temperature, time and duration of mechanical milling for

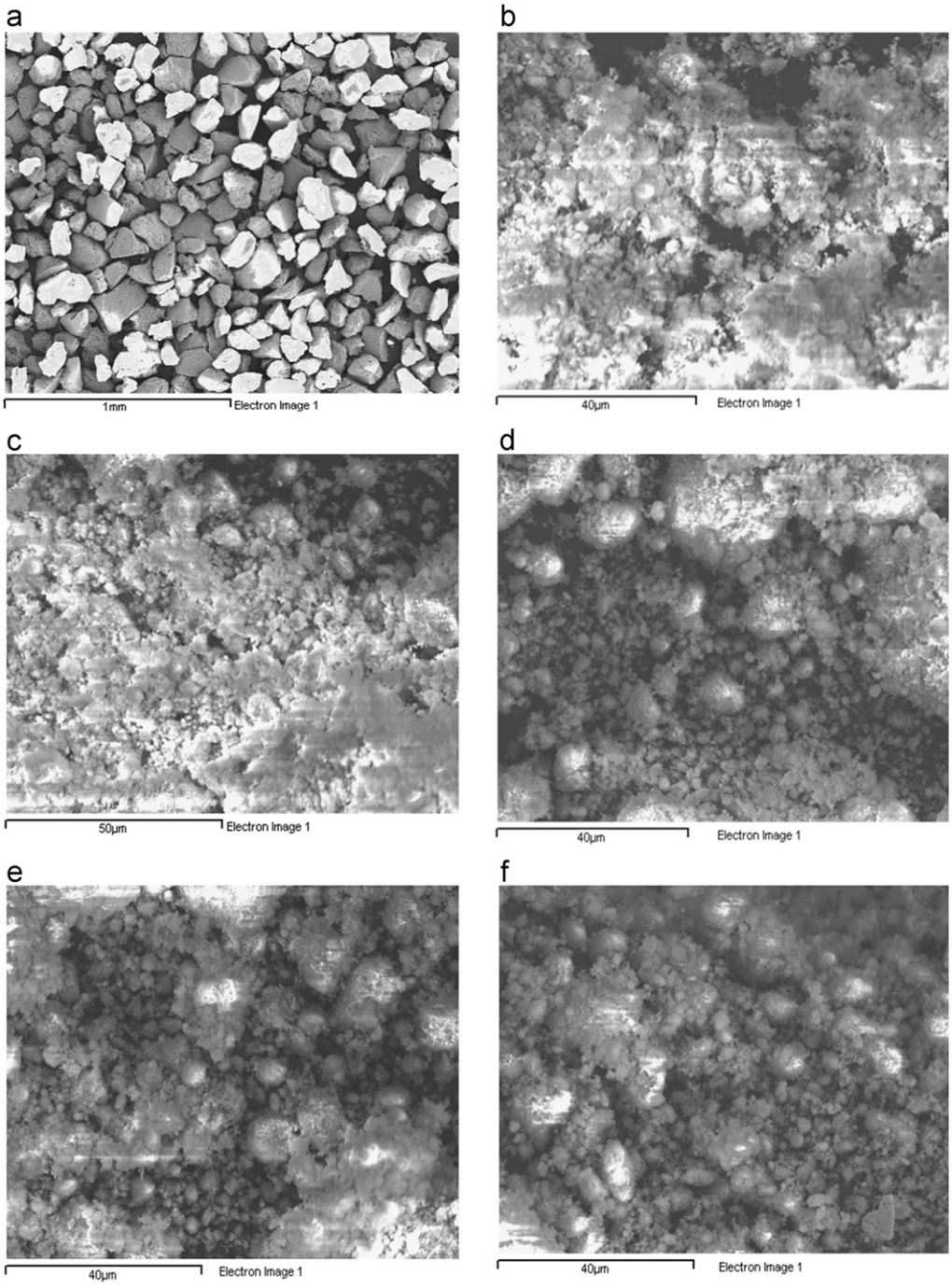

Fig. 3. The surface morphology of silica sand after undergoing mechanical treatment at various grinding times: (a) as-received, (b) 20 h, (c) 40 h, (d) 60 h, (e) 80 h and (f) 100 h.

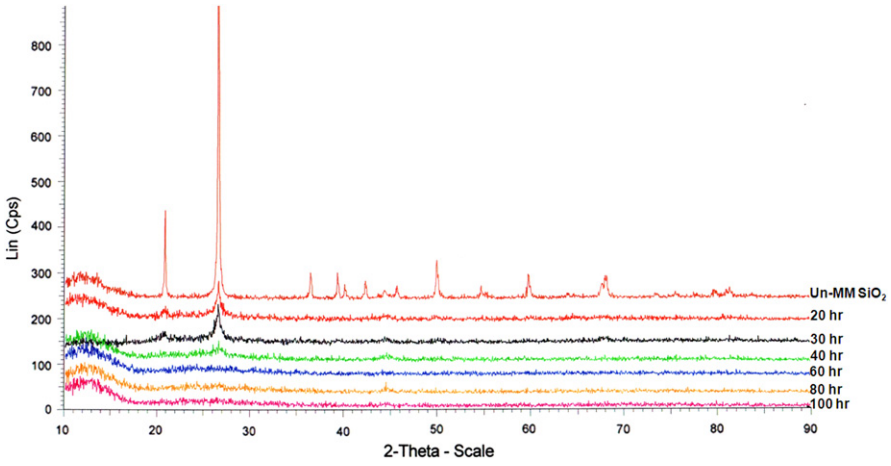

Fig. 4. XRD spectrum of mechanically milled silica at different milling time.

Table 9
Intensity value and d_{50} at various milling time.

Sample	Milling time (h)	Intensity (counts)	d_{50} (μm)	Random disorder (%)
Un-MM	0	800	64.00	
MM1	20	100	3.46	87.5
MM2	30	80	3.38	90.0
MM3	40	45	2.61	94.4
MM4	60	40	2.58	95.0
MM5	80	30	1.26	96.2
MM6	100	20	1.18	97.5

silica sand. These observations indicate that the percentage yield of SiC (in terms of the weight of product obtained) formation would increase when temperature, time, and duration of mechanical milling for silica sand increases. In contrast as the heating rate is increased, the percentage yield of SiC formation is lowered.

The formation of SiC depends on several factors. According to Ibrahim et al. and Kishore et al. [24,25] the combination effects of different factors can be determined by decomposition of the total variation into appropriate components known as analysis of variance (ANOVA). ANOVA is also needed for estimating the error variance. For the present study, the ANOVA results are given in Table 6.

Ibrahim et al. [24] reported that the variance of each factor is obtained by dividing the sum of squares for each factor with its DOF. They also mentioned that the degree of freedom associated with a factor equals to one less than the number of levels. According to them, for a factor with three levels, level 1 data can be compared with level 2 and level 3 data but not with level 1 itself. Thus, the three level factors have two DOF. The authors explained that the variance ratio (F) is the ratio of variance due to the effect of a factor to the variance owing to the error term.

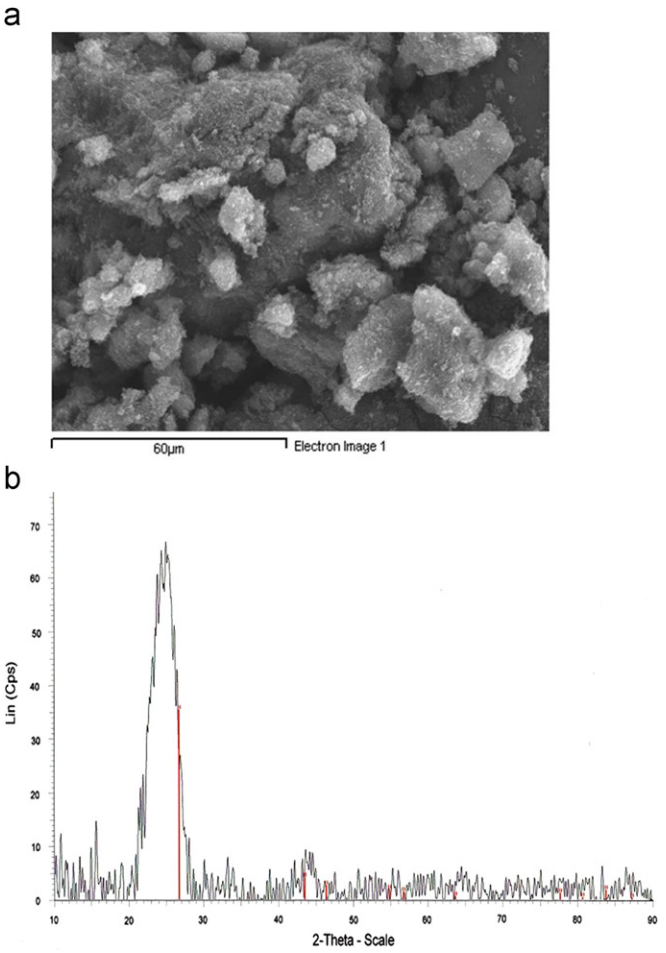

Fig. 5. (a) SEM micrograph of ball milled carbon and (b) XRD spectrum of ball milled carbon.

According to the data presented in Table 6, it was obvious that the factor duration of mechanical milling for silica sand contributed the highest percentage (55.83%), followed by temperature (22.12%), time (14.81%) and heating rate (7.21%). Since the contribution of heating rate is the smallest

(i.e. less than 10%), it is considered insignificant and this factor is pooled i.e. combined with the error term. Ibrahim et al. [24] described pooling as a process of ignoring the contribution of a selected factor and subsequently adjusting the contribution of the other factors. The new ANOVA result after pooling is as tabulated in Table 7.

As can be observed from Table 7, the percentage contribution of the remaining factors decreased slightly after pooling, but ranking of the factor effects still remained the same as before. The average performance of effects indicates that level 3 (see Table 5) for factors duration of mechanical milling for silica sand, temperature and time gives the optimal condition. As shown in Table 8, the expected improvement is 4.69 over the current average performance of 17.83. Since heating rate had little significance, it was omitted in the selection of levels for the optimum condition [24–28].

Based on these results, the highest % yield of SiC formation is attained at the combination setting of A_3, B_3, C_1 and D_3, i.e. processing temperature of 1650 °C, processing time of 180 min, heating rate of 5 °C min^{-1} and duration of mechanical milling for silica sand of 100 h. This combination of settings was not used in the experiments. According to Ji et al. [26] the orthogonal array is capable of identifying optimum factors in a multidimensional factor space.

The expected results ($R_{expected}$) at optimum condition in terms of S/N ratio can be converted back to the scale of units of the original observations as follows:

$$2.53 = -10\log(\mathrm{MSD})$$
$$\mathrm{MSD} = 10^{-22.53/10} = 5.53 \times 10^{-3}$$
$$\text{Therefore, } R_{expected} = (\mathrm{MSD})^{-1/2} = 13.387\ \mathrm{g}$$

Verification experiments were conducted by running another three replications at the combined setting of A_3, B_3, C_1 and D_3. The average yield of SiC was found to be 12.824 g which is within the confidence interval of the predicted optimal yield formation of SiC.

3.2. Silica sand and carbon characteristic

The surface morphology of the Un-MM and MM silica sand are shown in Fig. 1(a) and (b), respectively. The powder of Un-MM silica sand exhibited angular structure whereas the MM batch type had distorted structure after it underwent the mechanical milling process.

XRD analysis of the Un-MM silica sand shows the crystalline behaviour of the sample (see Fig. 2a). The diffraction line of silica can be clearly seen before mechanical milling process. In crystalline forms, the structure is characterised by tetrahedral configuration of atoms within the crystals. It shows discrete reflection in X-ray diffraction from

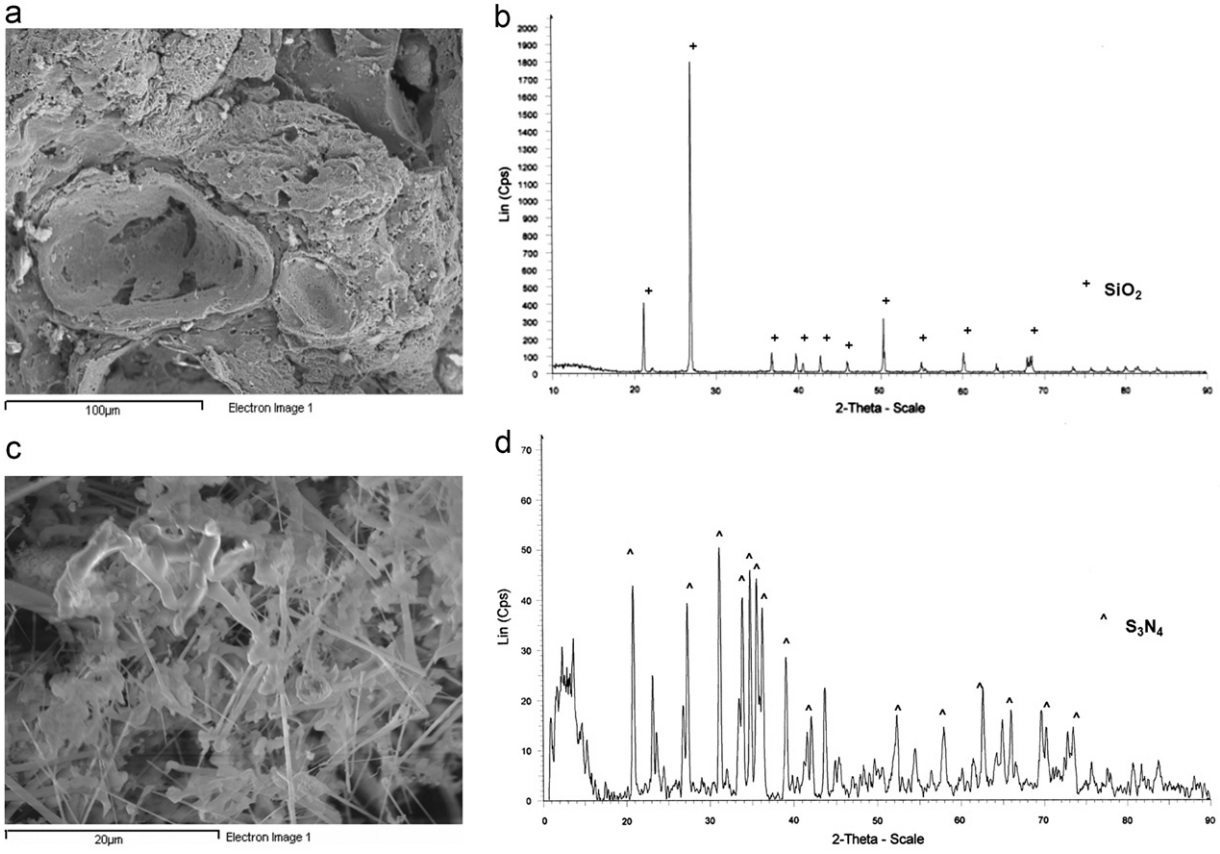

Fig. 6. (a) SEM micrograph of Un-MM mixture at 1350 °C, (b) XRD spectrum of Un-MM mixture at 1350 °C, (c) SEM micrograph of MM mixture at 1350 °C and (d) XRD spectrum of MM mixture at 1350 °C.

the internal planes formed by the orderly patterns of atoms. The mechanically milled silica however, exhibits crystal lattice distortion, showing the pattern of a vast broadening effect at the base of the peaks and also some reduction in intensity as shown in the spectrum of Fig. 2(b). The surface morphology of the silica sand subjected to various milling times is shown in Fig. 3(a–f).

The XRD patterns shown in Fig. 4 are for milled silica sand with different milling periods ranging from 0 to 100 h. The highest peak for Un-MM silica sand is at 26.2° (2θ angle) and this peak was taken as reference. The intensity of the peak after 20 h of milling was reduced to 87.5%. A great intensity reduction of up to 97.5% was observed after 100 h of milling. As particle size of (MM) silica sand reduces due to longer milling time, broadening effects at the peak base of the spectrum increases.

This observation is in agreement with the work of Suryanarayana [20] who found similar XRD pattern i.e. reduction of peak intensity and broadening effects for talc samples. Szewezak and Wyrzykowski [21] reported that the maximum size reduction limit for a particle is equivalent to the size of tough-brittle transition which leads to plastic deformation rather than breakage of particles and it is accompanied by growth of structure distortion.

Table 9 shows that the degree of random disorder in mechanical milling (MM) silica sand increases as milling time increases. The degree of random disorder is about 98% after a milling time of 100 h. This shows that the mechanical milling process which is based on impact and friction between particles not only causes size reduction but also produces important changes in the physio chemical properties of solids, and leads to structural alteration by loss of regularity in the crystalline network. An important method employed in investigation of distortion is the observation of line broadening in the X-Ray diffraction spectra. The intensity and half width of the X-Ray lines in different patterns reflect the degree of random disorder and lattice distortion of the particles.

According to Romero et al. [22] the main problems of the carbothermal nitridation process are the particle size of the reactants and the need for a close contact between them. In an attempt to avoid these problems, ball-milled carbon was used as a starting reactant. The carbon used here had undergone ball milling treatment for five hours.

The carbon powder exhibits angular and cubic shapes as shown in Fig. 5(a). This carbon powder has been studied by means of X-ray diffraction, which showed its amorphous structure as shown in Fig. 5(b).

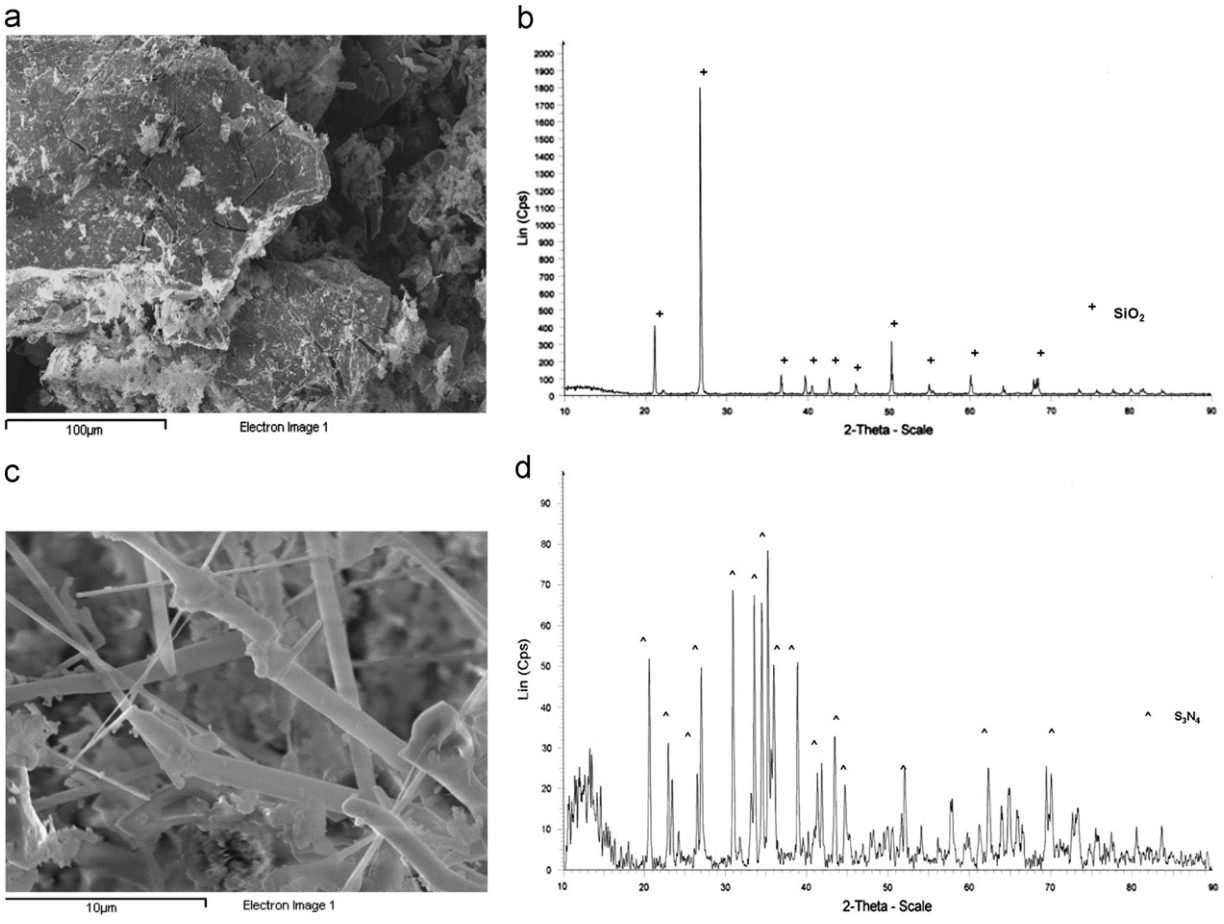

Fig. 7. (a) SEM micrograph of Un-MM mixture at 1450 °C, (b) XRD spectrum of Un-MM mixture at 1450 °C, (c) SEM micrograph of MM mixture at 1450 °C and (d) XRD spectrum of MM mixture at 1450 °C.

3.3. Carbothermal nitridation process of un-MM and MM silica sand at various temperatures

The surface morphology of the Un-MM mixture after the carbothermal nitridation process at 1350 °C is shown in Fig. 6(a). No formation of silicon nitride or silicon carbide compound was detected. The XRD spectrum of Fig. 6(b) indicated the presence of unreacted silica. The formation of silicon nitride was visible in the mixture of MM mixture at 1350 °C (Fig. 6c). At this temperature, the compound present was non-filamentary, although there were some primitive fibrous structures which were observed. The presence of silicon nitride compound was shown by XRD spectrum (Fig. 6d). This showed that the mechanical milling treatment on silica sand not only caused size reduction but also structural changes from the crystalline phase to a distorted structure. Silva and Figueiredo [5] explained that the formation of this compound is by means of a vapour–liquid–solid (VLS) mechanism. They also described that the VLS mechanism is characterised by the existence of impurity that acts as a preferred site for the deposition of gaseous intermediates that lead to supersaturation of liquid in the elements forming the crystal.

Fig. 7(a) shows the surface morphology of the Un-MM mixture at 1450 °C. The presences of silicon nitride is not observed and this is confirmed from the XRD spectrum as shown in Fig. 7(b). The crystalline nature of Un-MM silica sand makes it very stable even at 1450 °C and prohibits them from becoming gaseous intermediate in this reaction. According to Alcala et al. [16] this reaction occurs via a gas–solid mechanism, with silicon monoxide produced as an intermediate. The SEM micrograph shown in Fig. 7(c) indicated that the MM mixture has a homogenous composition constituting of the whisker shape compound of silicon nitride as confirmed by the XRD spectrum of Fig. 7(d). Alcala et al. [16] described that the formation of whisker silicon nitride could be associated with the formation of β-Si_3N_4. They reported that this crystal precipitated from the liquid at the liquid–solid interface, initiating a whisker that carried the particle from where it was formed at the top.

The surface morphology of the mixture after carbothermal nitridation process at 1550 °C is shown in Fig. 8(a). The presence of a spiky-like compound was observed. The XRD spectrum revealed there is no indication of silicon carbide formation in the structure as shown in Fig. 8(b). From the results obtained, formation of SiC was not favoured due to the crystallinity characteristic of the silica sand used in the

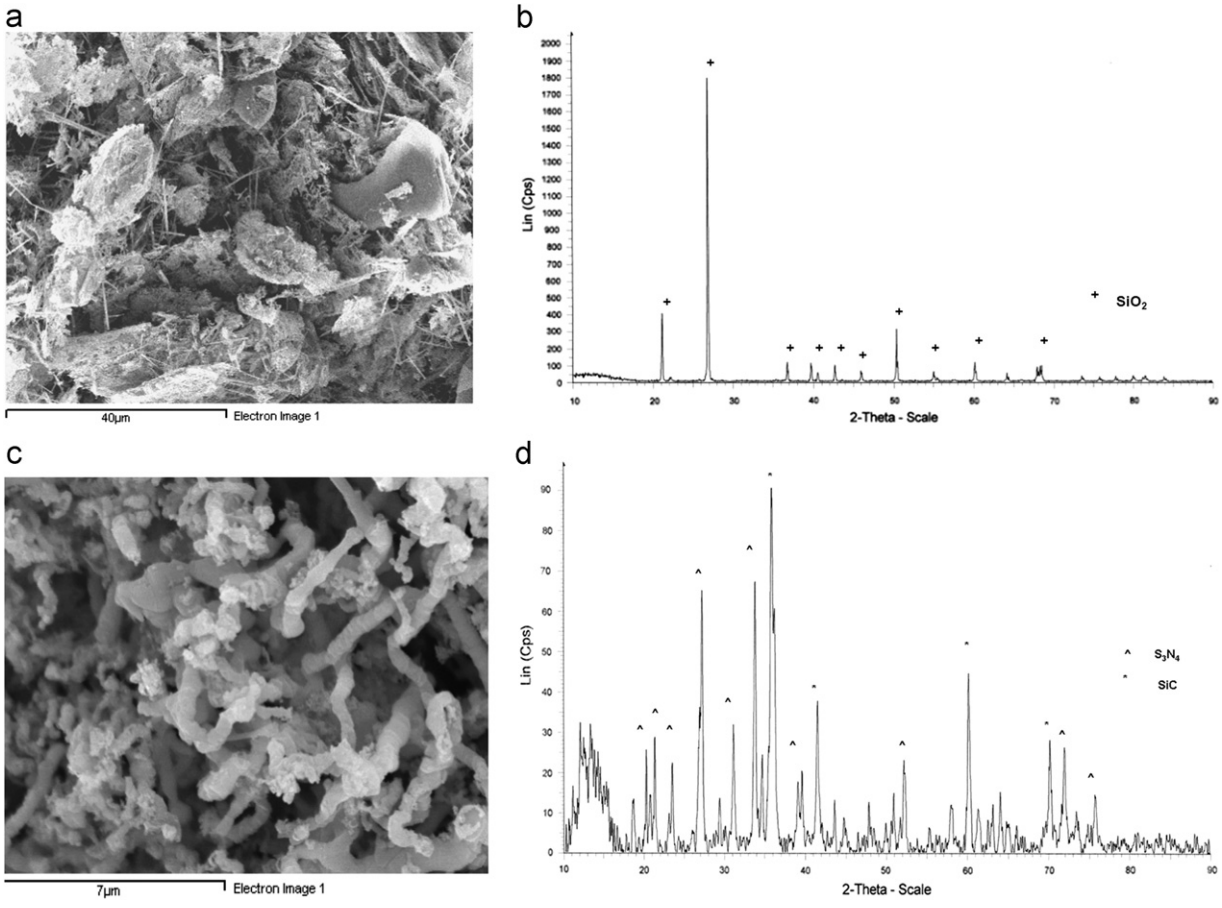

Fig. 8. (a) SEM micrograph of Un-MM mixture at 1550 °C, (b) XRD spectrum of Un-MM mixture at 1550 °C, (c) SEM micrograph of MM mixture at 1550 °C and (d) XRD spectrum of MM mixture at 1550 °C.

experiment. The morphology characteristics of the product of MM mixture obtained at this temperature (1550 °C) consisted of mainly silicon carbide whiskers and some silicon nitride compounds which were identified by X-ray diffraction analysis (see Fig. 8c and d). This means that, at this temperature the formation of silicon carbide is more favourable compared to silicon nitride, as predicted by thermodynamic calculations. The increase in temperature to 1550 °C favours silicon carbide formation and increases the yield to a larger extent.

The surface morphology of the mixture after carbothermal reduction at 1650 °C is shown in Fig. 9(a) and the presence of a spiky-like compound was observed. The XRD spectrum revealed no indication of either silicon carbide or silicon nitride formation in the structure as shown in Fig. 9(b). Romero et al. [22] reported that as SiC formation will dominantly be controlled by diffusion of silicon monoxide into the carbon structure, silica must firstly decompose into silicon monoxide vapour. As a result of this phenomenon, the formation could be facilitated by using silica sand with distorted structure with iron elements or compounds as effective agents for catalysis. The morphology characteristics of the product obtained at 1650 °C is dominated by the presence of silicon carbide whiskers (see Fig. 9c). At this higher temperature, formation of silicon carbide is more favourable than silicon nitride as shown by the XRD trace in Fig. 9(d).

With the presence of carbon, silicon nitride will convert to silicon carbide with the release of nitrogen gas according to reaction of Eq. (4) at high temperature:

$$Si_3N_4(s) + C(s) \rightarrow SiC(s) + 2N_2 \quad (4)$$

According to thermodynamic calculations, free energy (ΔG°) for the reaction of Eqn. (4) becomes negative at a temperature of 1450 °C therefore the formation of silicon carbide becomes favourable [22].

The following Table 10 and Fig. 10 summarise the effects of increasing the temperature on product composition and on reaction yields, for both Un-MM and MM mixtures. For the MM-mixture, an increase in temperature from 1350 to 1450 °C lead to an increase of silicon nitride formation by 51%. This shows that the increase in temperature favours the formation of silicon nitride and increases the reaction yield to a large extent. However, at this temperature, the formation of silicon carbide was also observed. The quantity of silicon carbide formed increased with the temperature increment. It is believed that silicon carbide is formed from the reaction of silicon nitride and

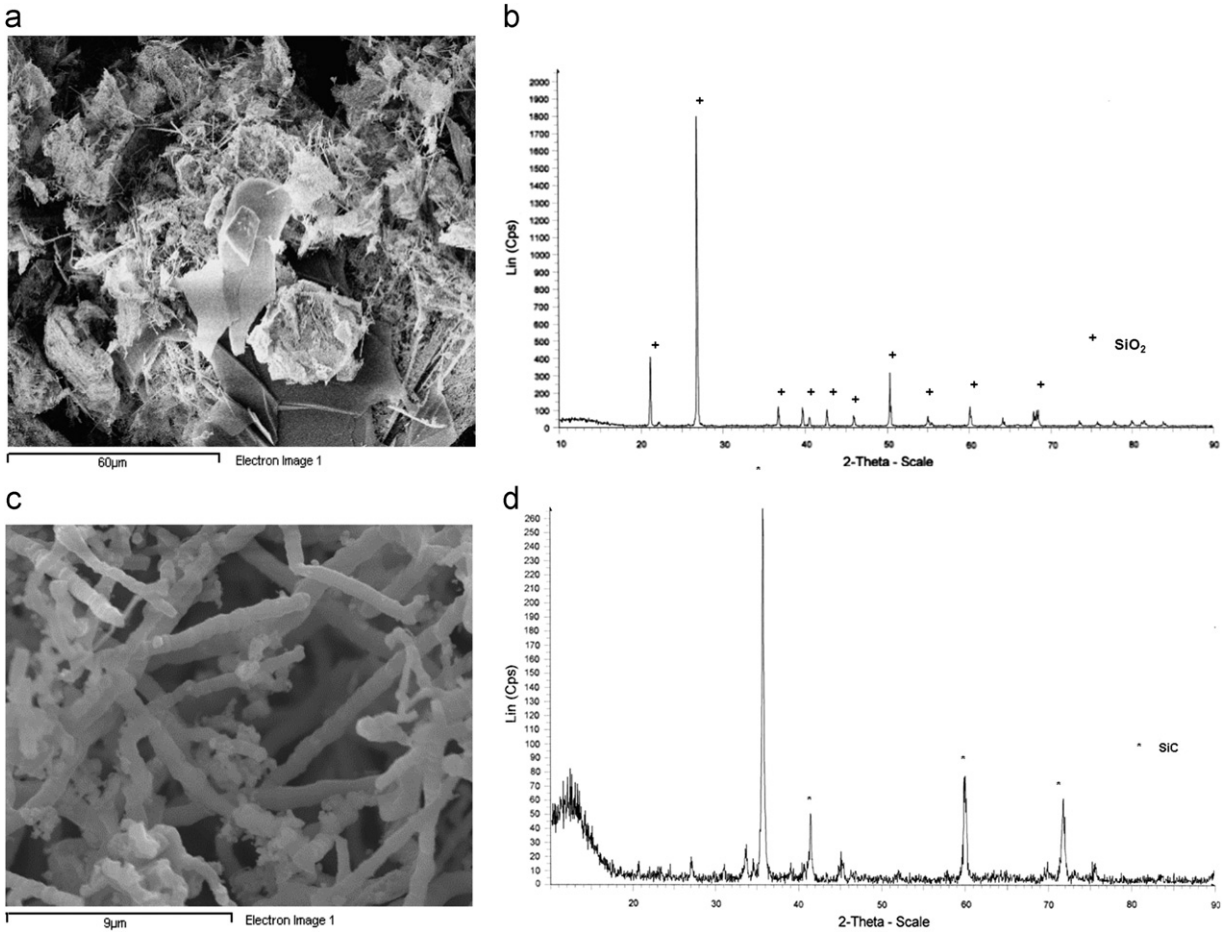

Fig. 9. (a) SEM micrograph of Un-MM mixture at 1650 °C, (b) XRD spectrum of Un-MM mixture at 1650 °C, (c) SEM micrograph of MM mixture at 1650 °C and (d) XRD spectrum of MM mixture at 1650 °C.

Table 10
The influence of temperature on the reaction yield and the product composition.

Temperature(°C)	MM-Mixture			Un-MM Mixture		
	% Yield	Si_3N_4 (%)	SiC (%)	% Yield	Si_3N_4 (%)	SiC (%)
1350	25.46	25.46	–	–	–	–
1450	82.59	76.48	6.11	–	–	–
1550	54.80	19.56	35.24	–	–	–
1650	74.72		74.72	1.54	–	1.54

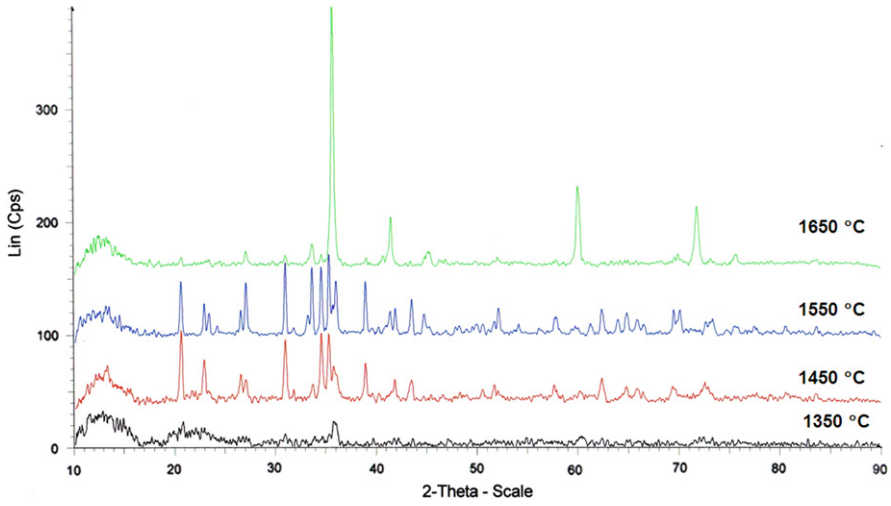

Fig. 10. X-ray diffraction pattern of mechanical milled silica and carbon mixture after carbothermal nitridation process at various temperatures.

carbon via the reaction mechanism of Eq. (4) as reported by Silva and Figueiredo [5].

4. Conclusions

In the present study, the influence of processing parameters on the yield formation of silicon carbide whiskers obtained from the carbothermal nitridation process using structurally altered silica sand have been optimised by employing Taguchi's Design of Experiments method and ANOVA. The optimal parameters and their corresponding levels were found to be A_3,B_3,C_1D_3, i.e., temperature of 1650 °C, time of 180 min, heating rate of 5 °C min^{-1} and duration of silica sand milling process of 100 h. The study showed that silicon carbide and silicon nitride whiskers were produced in silica sand and carbon solid mixtures in nitrogen-containing atmospheres. It was observed that the use of structurally distorted silica sand lowers the silicon carbide formation temperature and at the high process temperature of 1550 °C, the formation of silicon carbide whiskers was more favourable. The morphological characteristics of whiskers, which are of major importance for applications in composite materials, have been significantly improved by the use of structurally distorted silica sand in the carbothermal nitridation process.

Acknowledgement

The authors gratefully acknowledge the Ministry of Science Technology and Innovation Malaysia (MOSTI) for the financial support under the Grant no. SF03-03-02-SF0146.

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