# Interaction between silicon carbide and a nickel-based superalloy at elevated temperatures

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A study has been made of the reaction of hot-pressed SiC and a nickel-based superalloy at temperatures between 700 and 1150° C. Under conditions of reduced oxygen pressure at the reaction interface, obtained by applying pressure to the couple, some degree of reaction was observed in both metal and ceramic at all temperatures studied. Preliminary studies utilizing the same techniques at 1000° C with a Si–SiC ceramic composite, Si<sub>3</sub>N<sub>4</sub>, MgO, Al<sub>2</sub>O<sub>3</sub>, and SiO<sub>2</sub> also indicated some degree of reaction in the metal for all ceramics examined.

## 1. Introduction

Silicon-based ceramics are currently being investigated for use as structural components in hot machinery such as gas turbines. In these applications, ceramics and metals will be in contact with each other in areas where the interface temperature can be in excess of  $1000^{\circ}$  C. Furthermore, in the area of contact there is likely to be a tight fit between the metal and the ceramic. Within this area of intimate contact, oxygen will tend to be excluded, preventing the formation of an oxide layer on either metal or ceramic. It is appropriate to consider the possibility of ceramicmetal reactions under such conditions.

The subject of ceramic-metal solid-state reactions under inert or reducing conditions received considerable attention ten or fifteen years ago when research was conducted on metal matrix composites. The reinforcing filament consisted mainly of boron, BORSIC\* (boron coated with silicon carbide), silicon carbide, alumina, carbon, glass and various metal filaments. Much of this work has been summarized in two volumes dealing with interfaces and properties of metal matrix composites [1, 2]. It has been known since 1966, for example, that SiC filaments react in a

reducing environment with Fe, Co, and Ni at temperatures as low as 700° C [3]. The reactivity of SiC filaments with a number of nickel-based superalloys, as well as an Fe-Cr-A1-Y alloy, was recently reported by Cornie *et al.* [4]. All the alloys studied reacted to some extent with SiC during hot isostatic pressing in vacuum at 996° C, forming silicides in both metal and the filaments. Indeed, the extent and severity of these and similar reactions, as well as the difficulty in reducing them by protective coatings, has impeded the development of metal matrix composites using boron, BORSIC, SiC, or Al<sub>2</sub>O<sub>3</sub> filaments in any metal matrix with a melting point higher than Al.

Similar reactions in vacuum or inert gas have also been found between  $Si_3N_4$  coupons and V, Ti, Nb, and Ta at temperatures between 1200 and 1600° C [5]. Bennett and Houlton [6] have studied the interactions of  $Si_3N_4$  with several iron, nickel, and molybdenum alloys between 800 and 1000° C in inert gas and in all cases found some degree of a reaction between metal and ceramic.

For structural ceramics containing free silicon (e.g., Norton NC 435, UKAEA's self-bonded silicon carbide, General Electric's Si-SiC), Mehan and McKee [7] found severe reactions in air with

\*Trademark of United Aircraft Corp.

Ni, Co, and a nickel-based superalloy (IN-718) at temperatures above  $1000^{\circ}$  C. However, it was concluded that the reaction between IN-718 and both SiC and Si<sub>3</sub>N<sub>4</sub> was relatively benign at a temperature of  $1150^{\circ}$  C. In these experiments loosely coupled coupons were used, tied together with metal wire, which allowed oxide layers to be formed on both ceramic and metal. This was not sufficient to prevent silicon diffusion between a ceramic containing free silicon and the metal, but considerably reduced it for the case of SiC and Si<sub>3</sub>N<sub>4</sub>. In a vacuum, SiC reacted severely with IN-718 at 1150° C, in agreement with all previous work.

When considering the problem of metal-ceramic contact under conditions likely to be encountered in real machinery, and devising ways to study such reactions, it was concluded that experiments conducted in reducing or inert atmospheres were too severe and, in view of all past work, the results predictable. A system which brought the ceramicmetal coupons together in air and allowed an application of load to the assembly seemed to be the most realistic in simulating service conditions. It is the purpose of this paper to describe the results of a study employing such a system using SiC and a nickel-base superalloy as the reacting species. The study by necessity is phenomenological in nature, since a typical nickel-base superalloy contains about ten alloying elements of which at least six can form silicides and/or carbides. To sort out detailed mechanisms of the reaction of Si and C (the expected products of the decomposition of SiC) with such a material is quite difficult and probably unnecessary. What is needed from a practical viewpoint is a knowledge of the nature and severity of the reaction, identification of the reacting species, and if possible some phase identification. It is to these questions that this work is addressed.

## 2. Experimental

#### 2.1. Materials

The ceramic used was hot-pressed SiC manufactured by the Norton Company. In all cases but one the ceramic was used in the as-machined condition. The exception was when a coupon was heat treated to form a protective  $SiO_2$  layer on the surface to study the effect of this layer on the reaction process.

The superalloy used was a nickel-based alloy whose nominal composition is given in Table I. A 2472

TABLE I Nominal composition of nickel-based superalloy

Element	Nominal composition (wt%)	
С	0.17	
Cr	14.0	
Ni	60.0	
Co	9.5	
Мо	4.0	
W	4.0	
Ti	5.0	
Al	3.0	
В	0.015	
Zr	0.03	

Ni-based superalloy was chosen because they are widely used and can be expected to be employed in machinery in conjunction with ceramics. Although the Ni–Si binary phase diagram shows a lower liquidus temperature ( $964^{\circ}$  C) than either Co–Si or Fe–Si (the basis of other superalloys), this would not alone be expected to deter the use of Ni-based alloys because of their other advantages. Further, it was considered of interest to determine if the kinetics of the reaction changed above and below the liquidus temperature. In all cases, the metal was used in the as-heat-treated and machined condition.

#### 2.2. Reaction couples

The specimens were machined into coupons 6.35 mm in diameter and 3.18 mm high. Prior to assembly, the metal specimens were polished on 400 mesh SiC paper, the ceramic specimens on a  $30\,\mu\text{m}$  diamond wheel, and both were cleaned in acetone.

The method used to apply the load is illustrated in Fig. 1. An Instron machine was used in all the experiments because of its ability to maintain accurate alignment and apply a variety of loads (by operating the machine in its load cycling mode). Since then, a simpler dead weight loaded apparatus has been designed and built; future work will be performed using this facility.

Referring to Fig. 1, the load cell and upper holding fixture were aligned in a lathe by manipulating the four bolts that attach the holding fixture to the load cell. Concentricity with respect to the load cell centre-line was maintained to within 0.0254 mm. Prior to assembling the alumina rods, thermocouples, and specimen assembly, a 2.54 cm diameter bushing was slipped over the 2.54 cm diameter holding rod fixtures (slip fit), and the upper and lower fixtures were brought closely

Figure 1 Photograph of the experimental apparatus used in this investigation.



together by raising the cross-head to which the lower holding fixture was attached. The lower holding fixture was attached to the cross-head by a coupling (not shown in Fig. 1) free to move small distances in both the horizontal and vertical plane. This coupling was loose as the upper and lower fixtures were brought together and maintained in alignment by the bushing. The coupling was then tightened and the bushing was slipped back and forth between the two holding rod fixtures to make sure they were aligned. The cross-head was lowered, the bushing removed, and alumina rods (1.18 cm in diameter and extending 1.9 cm into the holding fixture), thermocouples, and reaction couple were inserted, and finally a 3.0 cm diameter tube furnace was positioned over the assembly.

The entire specimen and fixture train were designed to be concentric and in alignment to within 0.0254 mm. These tight tolerances were required to keep the metal-ceramic reaction interface flat and parallel in order to maintain uniform contact. In general, this objective was met, as judged by the uniformity of the reaction zone.

A stress level between 2.8 and  $7.0 \text{ MN m}^{-2}$  was maintained on the reaction couples. Lower stresses were used at the higher temperatures, and all reaction runs were carried out for 100 h. The effect of time on the depth of attack was not studied in this investigation. In the analysis of the reaction kinetics, it was assumed that the reaction zone thickness was a function of the square root of time (diffusion controlled). This point will be

examined in more detail subsequently; however, this assumption does not seem unreasonable as diffusion was found to be the rate controlling process in the metal— $Si_3N_4$  system [5], as well as the  $Ni-Al_2O_3$  system [8].

# 2.3. Methods of analysis

The reaction zones formed in these experiments were analysed by several techniques. After a reaction run was completed and the surfaces examined visually, the specimens were sectioned and examined by standard metallographic techniques. When microhardness data were required, a Knoop indenter with a 100 g load was employed. Scanning electron microscopy (SEM) and semiquantitative energy dispersive X-ray spectroscopy (EDS) were performed with an ISI model Super III equipped with a Nuclear semi-conductor detector and a Tracor Northern model 2000 analyser. X-ray spectra were recorded photographically and in some cases stored on magnetic discs for subsequent processing. Quantitative analyses (EMA) were done in a semi-automated Cameca model MS 46 microprobe equipped with four wavelength dispersive spectrometers. A complete analysis consisted of measuring characteristic X-ray intensities from four different elements on each of two successive data collection sequences. These included Ni, Cr, Si and W follwed by Co, Ti, Mo and Al. Carbon was accounted for by difference.

An operating voltage of 15 kV was selected for quantitative analysis as a compromise between minimum beam penetration and maximum signal intensity. A beam current of 45 nA and a counting interval of 30 sec resulted in sufficient counts to yield statistical minimum detectability limits between 0.1 and 0.5 wt % and relative precisions of 10% or better for most elements. The choice of WM $\alpha$  proved to be a problem due to a partial overlap of SiK $\alpha$ . This resulted in erroneously high values for W in the presence of high Si concentrations. X-ray data were first corrected for instrumental dead time, drift, background and then formed into "k" ratios (i.e., the intensity from the sample divided by the intensity from its corresponding pure element standard) These "k" ratios were subsequently converted into compositions using a modified version of a quantitative analysis computer program [9].

X-ray diffraction analysis (XRD) was used without difficulty in partially identifying the

reaction products formed on the SiC because the product was easily removed. However, because of the complexity of the reaction zone on the metal side, its strong adherence to the base metal, and its small relative concentration with respect to the unreacted material, XRD techniques were difficult to use and interpret. The technique finally adopted was to grind away carefully most of the unreacted material, leaving the reaction product in a relatively concentrated form. Analysis was still difficult because of the number of phases present, but several were identified.

# 3. Results

# 3.1. Metallography and microhardness

A reaction product was detected on both the superalloy and SiC at all temperatures studied between 700 and  $1150^{\circ}$  C. At  $700^{\circ}$  C, the reaction was light and discontinuous; at  $1150^{\circ}$  C the reactions on both metal and ceramic were severe. This is in marked contrast to the results reported previously [7], where protective oxide films are allowed to form and little reaction occurred.

Photomicrographs of the reaction zone formed in the superalloy at 900 and  $1150^{\circ}$  C are shown at the same magnification in Figs. 2 and 3. Fig. 4 shows the reaction zone formed at  $1050^{\circ}$  C at a higher magnification and with a light etch. At least three metallographically distinct phases are present, and undoubtedly more are present. The length and thickness of the "fingers" extending into the metal from the reaction interface decrease as the reaction temperature decreases and are absent at 900° C and below. However, careful examination again revealed that at least three phases were present.



Figure 2 Reaction zone in the nickel-base superalloy after reaction with SiC for 100 h at  $900^{\circ} \text{ C}$ .



Figure 3 Reaction zone in the nickel-base superalloy after reaction with SiC for 100 h at  $1150^{\circ}$  C.

Metallographically, the reaction zone in the SiC appeared the same irrespective of temperature. This is illustrated in Fig. 5, which represents the reaction zone formed at 1050° C. At all temperatures investigated, a banded structure was present and cracks were visible in the reaction zone and SiC base material. Assuming the reaction product contains some silicides (see Section 3.2), the cracks are probably due to the difference in thermal expansion between the silicides and the SiC. Most silicides of Ni, Ti, and Cr, for example, have coefficients of thermal expansion greater than  $8 \times 10^{-6} \,^{\circ} C^{-1}$  [10], as compared to about  $5.5 \times 10^{-6}$ ° C<sup>-1</sup> for SiC. Cracking could also have been introduced during sectioning of the specimen due to relief of the residual stress.

Microhardness measurements were taken in the reacted region of the alloy specimen after exposure to SiC at  $1050^{\circ}$  C and adjacent to it in the unreacted matrix. The indenter could not determine the hardness of individual phases because of



Figure 4 Reaction zone in the nickel-base superalloy after reaction with SiC for 100 h at  $1050^{\circ}$  C. Specimen lightly etched with HF/HNO<sub>3</sub> lactic acid.



Figure 5 Reaction zone in SiC after reaction with nickelbase superalloy for 100 h at  $1050^{\circ} \text{ C}$ .

its size, so an averaged value of hardness was obtained. In the reacted area, Knoop hardness values between 1020 and 1140 were observed; in the unreacted base metal, an average Knoop hardness of 397 was measured. The hardness values in the reacted region are not inconsistent with silicide formation (TiSi, 1039; NiSi<sub>2</sub>, 1019; CrSi<sub>2</sub>, 1131), but are low for carbides (TiC, 2470; Cr<sub>23</sub>Cr<sub>6</sub>, 1650) [10]. A hardness value of 397 is reasonable for a superalloy subjected to a 100 h exposure at a temperature of 1050° C.

Two experiments were conducted to determine whether (a) the presence of a thin  $SiO_2$  layer on the SiC affected the degree of reaction and (b) increasing the stress had an effect. In the preoxidation experiment, the SiC coupon was heated 16 h at 1200° C to form the SiO<sub>2</sub> layer which provides the oxidation protection for this class of materials. A 100 h reactivity experiment with the as-machined superalloy was conducted at 1100°C, and no difference in reaction kinetics or metallographic appearance was detected on either metal or ceramic. The effect of load was studied at 800°C, and the difference between a stress of  $28.1 \text{ MN m}^{-2}$  versus  $7.0 \text{ MN m}^{-2}$  was determined. The higher load gave a slightly more uniform reaction zone, but the reaction depth in both metal and ceramic was unchanged.

#### 3.2. Qualitative and quantitative analysis

Most of the analytical data were performed on specimens reacted at the highest temperature studied,  $1150^{\circ}$  C. At this temperature the reaction zone was largest which facilitated the use of EDS and EMA techniques. Sufficient work was performed at a lower temperature (900° C) to confirm the metallographic observation that the reaction zones were similar in kind but smaller. Similarly, much of the work was done on the metal side of the reaction couple because of its comlexity in comparison to the SiC side.

Fig. 6 illustrates the diversity of the reaction products formed by the diffusion of C and Si into the metal half of the couple at 1150°C. The secondary electron SEM micrograph reveals a minimum of seven separate phases, most of which are too small for quantitative EMA analysis. In general, beam penetration through the phase of interest, beam tailing and other sources of unwanted X-ray spectral artifacts [11] limit in situ analyses to structures five or more  $\mu m$  in size. It is possible, however, to determine relative differences between phases much smaller than this limit. Fig. 7 shows the semi-quantitative EDS spectra obtained from the points labelled A, B, and C in Fig. 6. Region A shows that Ni, Si, and Cr are predominant, and based on XDR data may be (Ni, Co) 16 Cr<sub>6</sub>Si<sub>7</sub>. Region B shows significant amounts of Ti and Mo in addition to Ni, Co, Cr, and Si, and probably consists of a single or several complex silicides\*. Particle C, because of its morphology and elemental content, may well be an  $M_6C$  carbide, which can be formed in this class of alloys after exposure to high temperature. Although it has been found that Si can enter into such a carbide, giving compositions such as (Ni, Co, Si)<sub>5</sub> (W, Cr)C [12], there are insufficient data to verify such a composition in this carbide.



Figure 6 Secondary electron SEM microphotograph of nickel-base superalloy reacted at  $1150^{\circ}$  C.

Quantitative EMA analyses were performed on the same specimen using the step scan mode in which the sample was translated  $10 \,\mu m$  between each reading. Fig. 8 is an optical micrograph of the analysed region showing the points analysed. Unfortunately, backlash in the specimen stage gave



Figure 7 Semi-quantitative EDS spectra from indicated areas in Fig. 6. The scale on spectra from point C is twice that of points A and B.

\*It was mentioned earlier (Section 2.3) that there is an overlap problem between WM $\alpha$  and SiK $\alpha$ . Low levels of W can be confirmed by the appearance of its higher energy L series emission lines. Although moderate to high concentrations of Si can be verified by a careful comparison of the relative M and L peak lengths, low concentrations are difficult to confirm by this technique.



Figure 8 Optical micrograph of EMA analysed area for the nickel-base superalloy reacted at  $1150^{\circ}$  C. Measured points have been enhanced by inking.

rise to a  $4\,\mu\text{m}$  misregistration between scans. In spite of this limitation, the resulting data are sufficient to determine the diffusion depth of the Si and the composition of the larger phases. Fig. 9 contains the computed weight fraction profiles for six of the eight measured elements. Examination of the Si profiles shows a small Si rich phase at  $30 \,\mu$ m from the edge, corresponding to point B in Fig. 6. A homogeneous phase containing 4.7 wt% is present about  $60 \,\mu$ m from the specimen edge extending about  $30 \,\mu$ m into the sample. It is associated with a high Ti and Mo content and could be a silicide of the form  $\Gamma i_{0.8} M_{0.2} Si_2$  [13].

Turning now to X-ray diffraction data obtained on the nickel-based alloy reaction zone formed at the same temperature (1150° C), only two phases could be positively identified; Ni<sub>16</sub> Cr<sub>6</sub>Si<sub>7</sub> and TiO<sub>2</sub>. The jagged dark band appearing about 50  $\mu$ m from the surface in Fig. 8 is high in Ti, and could be the source of the rutile. Point A in Fig. 6 is high in Ni, Cr, and Si and could well be the silicide region identified by XDR.

The reaction zone on the superalloy side at  $900^{\circ}$  C, which is below the Ni–Si eutectic, is much smaller than that present at  $1150^{\circ}$  C and it is not possible to separate the phases. EDS analysis shows that Si is present, as well as the same



Figure 9 Computed weight fraction for six of eight EMA analysed elements in the nickel-base superalloy.

elements found at  $1150^{\circ}$  C. Two spectra are shown in Fig. 10, one taken near the reaction zone surface and the other about halfway through the zone.

On the SiC side of the reaction couple, it has been pointed out that a banded structure is present at all temperatures studied. X-ray diffraction analysis at temperatures above 1000° C (none were obtained below this temperature) revealed the presence of a  $\delta$ -Ni<sub>2</sub>Si structure. EDS' analysis of the reaction zone revealed the presence of Cr, Co, W, and Al as well as Ni and Si at both 900 and 1150° C. An EMA 10 $\mu$ m step scan at 1150° C also showed an elevated Ti content. Evidently, the constituents of the superalloy are diffusing into the SiC, while at the same time Si and C are diffusing into the alloy.



POINT 3



Figure 10 Semi-quantitative EDS spectra for the nickelbase superalloy reacted with SiC at  $900^{\circ}$  C. Point 3 (top photograph) is near the surface and point 7 near the middle of the reaction zone. The scale on point 7 is twice that of point 3.

# 3.3. Reaction kinetics

Using a light etch as in Fig. 4, the reaction zone thickness on the superalloy could readily be measured. On the SiC, the reaction zone is well defined and needed no etching. Assuming, as mentioned earlier, that the reaction is diffusion controlled, the logarithm of the reaction rate constant (reaction distance divided by the square root of the exposure period [100 h]) may be plotted as a function of reciprocal of absolute temperature for both the metal and ceramic, and the plot shown in Fig. 11 is obtained. For the metal, a straight line seems to fit the data over the entire temperature range, yielding an activation energy of  $27.1 \text{ kcal mol}^{-1}$ . If the reaction is diffusion controlled, and because the diffusion coefficient is proportional to  $x^2/t$  (x being the diffusion distance) rather than  $x/\sqrt{t}$ , the activation energy for diffusion would be twice that obtained from the square root of time plot or 54.2 kcal  $mol^{-1}$ .

Below 900° C, the depth of the reaction zone in the SiC is the same as in the metal. Above 900° C, however, SiC reacts at a somewhat lower rate  $(Q = 2 \times 15.1 = 30.2 \text{ kcal mol}^{-1})$ , and perhaps not in a linear manner. The reason for this behaviour may be related to the liquidus at about 964° C in the Ni–Si system, although why this should slow down the reaction rate in the ceramic (and not affect that in the metal) is not immediately obvious.

## 4. Discussion

Based on the results presented in the previous section, it is evident that SiC undergoes an interchange reaction with the nickel-base superalloy. Si and C diffuse into the metal, and alloying constituents diffuse into the ceramic. Nickel is the predominant species diffusing into the ceramic, although Al, Cr, Co, Ti, and W are also present. Silicides are found in the metal; their presence lies predominantly near the surface as shown by the Si concentration profile in Fig. 9. Because C may be expected to diffuse more rapidly into the metal than Si, much of the reaction zone may consist of carbides. The fact that the microhardness data yielded values lower than typical carbide values may be a reflection of the fact that the size of the indenter yielded average values.

It is clear that the reaction process in this system is exceedingly complex because of the number of diffusing species. Nevertheless, it is of

TABLE II Reaction rate (cm sec <sup>-1/2</sup> )	at indicated	temperature
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Material	Data source	Reaction temperature (°C)		
		996	1093	
SiC	[4] *	$1.3 - 2.6 \times 10^{-5}$	$2.4 - 4.2 \times 10^{-5}$	
	Present data	$1.1 \times 10^{-5}$	$1.7 \times 10^{-5}$	
Ni alloy†	Present data	1.4 × 10 <sup>-5</sup>	$3.08 \times 10^{-5}$	

\*Two-hour reaction time.

†Metal reaction depth not available from [4].

some interest to compare the present results with those reported in [4]. In that investigation, the SiC filaments reacted with various matrices during fabrication to form silicides that metallographically appeared similar to the reaction product formed on the SiC in this investigation. The comparison between the two sets of data is shown in Table II. As may be seen, the data in [4], covering a variety of alloys, fall into the same range of reaction rates as the present data.

It is also possible to make a rough estimate of the difference in reaction rates when  $Si_3N_4$  is involved, rather than SiC. Bennett and Houlton [6] studied a number of alloys in contact with  $Si_3N_4$  in an inert gas but under no pressure. One alloy studied was Hastalloy X, which also was used in [4]. For the data of Cornie *et al.*, a 2 h exposure

at 996°C resulted in a reaction depth of  $18 \times 10^{-4}$  cm into the fibre, giving a reaction rate of  $2.1 \times 10^{-5}$  cm sec<sup>-1/2</sup> and is of the same order as observed in the present investigation (see Table II). In the case of  $Si_3N_4$ , after 1173 h in contact with Hastalloy X at 1000° C, a reaction depth of  $56 \times 10^{-4}$  cm was observed in the metal and  $16\times 10^{-4}\;\text{cm}$  in the  $Si_3N_4,$  resulting in reaction rates of  $2.7 \times 10^{-6}$  cm sec<sup>-1/2</sup> and  $7.8 \times 10^{-7}$  cm  $\sec^{-1/2}$ , respectively. These rates are an order of magnitude less than those observed in [4] and in the present work. A preliminary experiment performed in this laboratory involving hot-pressed Si<sub>3</sub>N<sub>4</sub> in contact with the same nickel-based superalloy for 100 h at 1000° C yielded a reaction rate of  $6.3 \times 10^{-6}$  cm sec<sup>-1/2</sup>, which is of the same order as that observed by Bennett and Houlton.



Figure 11 Plot of reaction rate and thickness for the nickel-base superalloy and SiC as a function of reciprocal of absolute temperature.

Moreover, the reaction morphology was completely different for the  $Si_3N_4$  case. In the metal a needle-like phase similar to that observed earlier by Mehan and McKee was found [7], while metallographically no evidence of a reaction was found on the ceramic. No analytical data have been obtained on this reaction couple, although more work is planned for a metal— $Si_3N_4$  system. It is probable that the reaction of  $Si_3N_4$  with metals is more complex than with SiC, because it has been shown that the partial pressure of  $N_2(g)$ plays a role in the former case [14].

The reaction between ceramics and nickel-base superalloys is not restricted to silicon-based ceramics. Using the experimental techniques described in Section 2 (but utilizing the simpler deadweight loaded apparatus referred to), preliminary experiments have been conducted at 1000° C. with the nickel-base alloy used in this investigation in contact with a free silicon containing ceramic (General Electric's Si-SiC\*), single crystal MgO,  $Al_2O_3$  and  $SiO_2$  (in the form of fused quartz). In all cases some degree of reaction occurred. In decreasing order of severity, Si-SiC showed the most severe reaction, followed by SiC,  $Si_3N_4$ ,  $Al_2O_3$ , MgO, and SiO<sub>2</sub>. In all cases a reaction product was present on the metal, but metallographically it was difficult to detect a reaction zone on the ceramic side for the last four materials.

In the case of the structural silicon-based ceramics, and to a lesser extent the oxide ceramics examined, it seems clear that applying a uniform pressure to the ceramic—metal interface while at temperature gives rise to reaction kinetics very similar to those obtained under vacuum or inert conditions. Based on past work performed on ceramic fibre-reinforced metal composites, it is clear that a protective coating is needed on the ceramic, the metal, or both. Also based on past experience, finding an impervious coating stable under conditions of cyclic temperature and extended time is not a trivial task.

## 5. Conclusions

From the results of this study, the following conclusions may be drawn:

(1) Hot-pressed SiC reacts with a nickel-base superalloy at all temperatures between 700 and

 $1150^{\circ}$  C under conditions where the presence of oxygen at the interface is minimized.

(2) The reaction seems to be diffusion controlled. For the metal, a straight line relation between reaction rate and the reciprocal absolute temperature is obtained with an activation energy of 27.1 kcal mol<sup>-1</sup>. On the ceramic side, the degree of reaction is the same as the metal below 900° C, but less above 900° C.

(3) The reaction mechanisms are quite complex, consisting of Si and C diffusing into the metal to form silicides and carbides, and the constituents of the alloy diffusing into the ceramic.

(4) Some degree of reaction at  $1000^{\circ}$  C was found under similar experimental conditions with  $Si_3N_4$ , MgO,  $Al_2O_3$ , and  $SiO_2$ .

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#### References

- 1. A. G. METCALFE, editor, "Interface in Metal Matrix Composites" (Academic Press, New York, 1974).
- 2. K. G. KREIDER, editor, "Metallic Matrix Composites" (Academic Press, New York, 1974).
- 3. R. F. SCHNEIDMILLER and J. E. WHITE, "Advanced Fibrous Reinforced Composites" Society of Aerospace Material and Process Engineering (SAMPE) 10 (1966) E-53.
- J. A. CORNIE, C. S. COOK and C. A. ANDERSON, "Fabrication Process Development of SiC/Superalloy Composite Sheet for Exhaust System Components", NASA-CR-134958 (1976).
- 5. P. LAMPARTER, S. STUB and A. GUKELBERGER, High Temp.-High Press. 3 (1971) 727.
- 6. M. J. BENNETT and M. R. HOULTON, J. Mater. Sci., 14 (1979) 184.
- 7. R. L. MEHAN and D. W. McKEE, *ibid.* 11 (1976) 1009.
- 8. R. L. MEHAN, Met. Trans. 3 (1972) 897.
- H. YAKOWITZ, R. L. MYKLEBUST and K. F. J. HEINRICH, "FRAME: An Online Correction Procedure for Quantitative Electron Probe Microanalysis", NBS Technical Note 796 (U.S. Government Printing Office, Washington, DC, 1973).

\*This material would be expected to be representative of any ceramic containing free silicon, such as Norton NC432 or the British REFEL material.

- G. V. SAMANOV, "Plenum Press Handbook of High-Temperature Materials – Property Index" (Plenum Press, New York, 1964).
- 11. R. B. BOLON and M. D. McCONNELL, in "Scanning Electron Microscopy 1976", edited by O. Johari (Illinois Institute of Technology Research Institute (IITRI), Chicago, Illinois, 1976) pp. 163-70.
- 12. C. T. SIMS and W. C. HAGEL, editors, "The Superalloys" (Wiley, New York, 1972) p. 55.
- 13. 1978 Powder Diffraction File, Publication SMA-28, International Center for Diffraction Data, Swarthmore, PA, Card 7-331.
- 14. E. H. ANDREWS, W. BONFIELD, C. K. L. DAVIES and A. J. MARKHAM, J. Mater. Sci. 7 (1972) 1003.

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