



HAL
open science

INTERFACES BETWEEN SiC AND METALS

M. Backhaus-Ricoult

► **To cite this version:**

M. Backhaus-Ricoult. INTERFACES BETWEEN SiC AND METALS. Journal de Physique Colloques, 1990, 51 (C1), pp.C1-769-C1-774. 10.1051/jphyscol:19901120 . jpa-00230029

HAL Id: jpa-00230029

<https://hal.science/jpa-00230029>

Submitted on 4 Feb 2008

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

INTERFACES BETWEEN SiC AND METALS

M. BACKHAUS-RICOULT

*Laboratoire de Physique des Matériaux, CNRS, 1, Place A. Briand,
F-92190 Meudon, France*

Résumé - Une étude des réactions chimiques se produisant aux interfaces entre métal et carbure de silicium a été effectuée pour divers métaux (Fe, Ni, Cr) et leurs alliages. Les chemins de réaction, les cinétiques de réaction, la morphologie macroscopique et la microstructure de la couche de réaction ont été déterminés. Un modèle a été développé pour expliquer l'influence de chaque composant métallique sur la composition et la structure de la couche de réaction.

Abstract - Chemical reactions occurring at metal-silicon carbide interfaces have been investigated for different metals (Fe, Ni, Cr) and their alloys. Reaction paths, reaction kinetics, macroscopic morphology and microstructure of the product scale have been determined. A model has been developed to explain the influence of each metal component on the composition and structure of the product scale.

1 - INTRODUCTION

Silicon carbide can be joined by diffusion welding under vacuum at high temperature. Chemical reactions occur at the metal-ceramic interface. Not only diffusion zones, but new products such as various silicides, carbides and/or free carbon have been observed in the interface region /1,2,3/. Only few fundamental studies can be found in the literature, for which the reaction products (structure and composition) and the reaction kinetics have been determined. Results on the possible products and the kinetics for the reaction of pure nickel or pure iron with silicon carbide are given in /4/, but no details on the microstructure are mentioned. More complicated alloys are studied in /1/, the microstructure has been analysed and seems to be very complex. To evaluate the influence of the different metallic compounds in steel-silicon carbide compounds we have performed careful experiments with pure metals (Fe, Ni), binary alloys (Fe-Ni, Fe-Cr, Ni-Cr) and finally ternary alloys or commercial steel (Fe-Ni-Cr). To obtain detailed information on the interface region we have determined the complete reaction path with all reaction products and diffusion zones, as well as the macroscopic morphology and the microstructure of the product scale.

2 - EXPERIMENTAL PROCEDURE

Various alloys ($Fe_{.75}Ni_{.25}$, $Fe_{.5}Ni_{.5}$, $Fe_{.25}Ni_{.75}$, $Fe_{.82}Cr_{.18}$, FeNiCr) were prepared by melting powder mixtures of pure materials obtained from Johnson Matthey (99.999). The homogeneity of the cylinders after annealing was controlled by microprobe analysis. The alloy and pure metal (Fe, Ni) cylinders were sawn into approximately 8 mm diameter, 2-3 mm thick disks. These disks were then polished with 100 grit SiC paper, followed by diamond paste down to 2 μm . The disks were finally cleaned in acetone. Polycrystalline silicon carbide with small impurity content (from Céramique et Composite) hot pressed pure silicon carbide and silicone carbide single crystals were cut into platelets and fine-polished with 2 μm diamond paste on a polishing wheel. Diffusion couples were formed by pressing ceramic platelet and metal disk against each other with the help of a graphite spring in a graphite holder to improve interfacial contact. The reaction temperature was measured by a thermocouple located near the reaction sandwich. The reactions were run under inert argon atmosphere in a graphite furnace at 1223 K for 1 to 100 hours. At the end of this period, the samples were quenched within a few minutes to room temperature, cut perpendicularly to the initial interface and polished by standard metallographic techniques. The polished-cross sections were investigated by optical and scanning electron microscopy. Concentration profiles of Si, C, Fe, Ni or Cr were measured with an electron microprobe. The composition

and the structure of the phases were also determined by X-ray diffraction. Thin foils were prepared by further polishing followed by ion milling with Ar at 5 KeV. The thin areas of the samples were investigated by transmission electron microscopy (Jeol 2000 FX) to obtain details on the microstructure.

Silicon carbide-iron

At 950°C, a thin, very irregular reaction layer could be detected, which consists of a silicon enriched α -iron matrix with embedded fibrous graphite particles (Fig.1). The reaction layer thickness varies strongly over the whole interface and does not increase systematically with time : reaction times of 2 hours as well as 100 hours yield a similar product scale thickness of about 10 μm . After reaction, systematic cracking at the metal/product scale interface was observed, whereas the SiC-product scale interface remained uncracked. The total reaction path is schematically represented in the quaternary Fe-Ni-Si-C phase diagram in Fig.2.

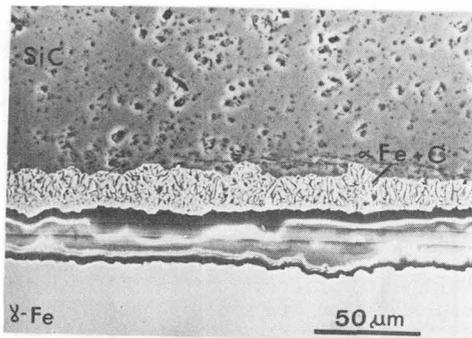


Fig.1 - Backscattered scanning electron micrograph of the reaction layer for the reaction of iron with silicon carbide at 1223 K.

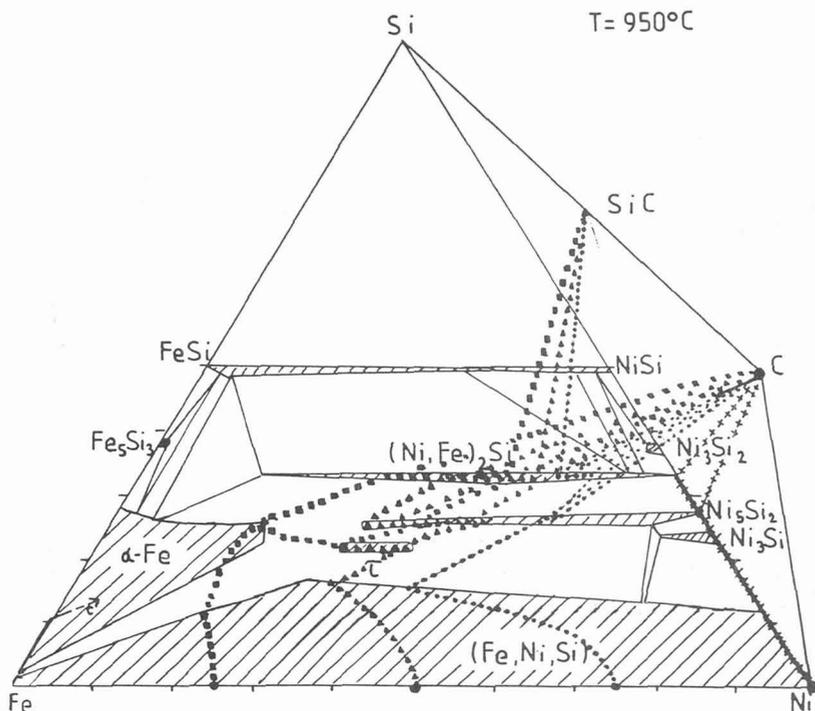


Fig.2 - Reaction paths for the solid state reactions between silicon carbide and different (Fe,Ni) alloys plotted into the quaternary Fe-Ni-Si-C phase diagram for 1223 K. (The reaction path is constituted by all the different compositions, which occur in the reaction sandwich. Therefore it represents a time-less diffusion profile in the four dimensional space of the phase diagram).

Silicon carbide-nickel

At 950°C, the reaction zone between nickel and silicon carbide is well organized in form of bands of constant thickness (approximately 10 μm) (Fig.3). Precipitate-free bands alternate with bands of the same matrix phase containing a high density of very small (~ 1 μm) graphite particles. The matrix composition varies from Ni₂Si on the SiC side, through Ni₅Si₂, to Ni₃Si, where this last composition forms only a precipitate-free zone next to the diffusion zone of the γ-nickel. The γ-phase is enriched in carbon and silicon. The total reaction layer thickness has been measured for various reaction times and has been found to grow with \sqrt{t} , with a parabolic rate constant $k = 1.25 \cdot 10^{-7} \text{cm}^2/\text{s}$. No cracking of the interface has been detected : cracks due to residual stresses during cooling occurs always within the ceramic.

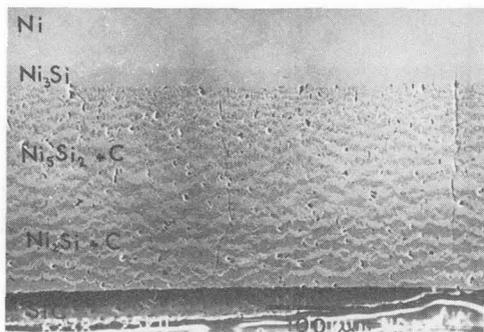


Fig.3 - Backscattered scanning electron micrograph of the reaction layer for the reaction of nickel with silicon carbide at 1223 K.

Silicon carbide-(iron, nickel, chromium) alloys

The morphology and composition of the product scale resulting from the reaction between silicon carbide and (iron, nickel) alloys always show similarities with those described above for pure metals. The strong regular banding of nickel-rich $(\text{Fe,Ni})_2\text{Si}$ (or $(\text{Fe,Ni})\text{Si}$) and $(\text{Fe,Ni})_2\text{Si}$ (or $(\text{Fe,Ni})\text{Si}$) + C, which is still visible for $\text{Fe}_{.25}\text{Ni}_{.75}$ evolves into a less regular structure for $\text{Fe}_{.50}\text{Ni}_{.50}$, where the bands are less organized and are already followed by a random distribution of precipitates near the metal side of the product scale. Finally, for $\text{Fe}_{.75}\text{Ni}_{.25}$ only a thin zone of these silicide bands can be detected; the major part of the product scale consists of τ -phase or silicon enriched α -Fe phase as matrix material with randomly distributed carbon precipitates. Transmission electron microscopy investigations of the product scale have proved, that the carbon precipitates have a fibrous character and consist of bundles of very well aligned turbostratic carbon. Energy dispersive X-ray microanalysis in a scanning transmission electron microscope did not show any special segregation of impurities in the precipitate interfaces. The matrix grains have a size close to or less than 1 μm. In the case of the reaction couple SiC- $\text{Fe}_{.75}\text{Ni}_{.25}$ a coexistence of α - and τ -phase together with carbon has been detected. For the cubic α -phase ($a = 5.75 \text{ \AA}$) ordering of the lattice (half lattice spacing) occurs in the [100] direction axis often only slightly misoriented, the slight tilt around the [111] axis or the twinning can be results of the diffusion controlled product scale growth.

An addition of chromium to the alloy does not influence the typical scale morphology, which is given by the iron and nickel content, but yields in addition a zone with chromium carbide precipitates on the metal side of the product scale (Fig.5,6). Different chromium carbides have been observed in coexistence in this precipitate region.

TABLE 1

Fe	(Fe _{.75} Ni _{.25})	(Fe _{.50} Ni _{.50})	(Fe _{.25} Ni _{.75})	Ni
γ -Fe	γ -alloy	γ -alloy	γ -alloy	γ -Ni
α -Fe	α -Fe			Ni ₃ Si
	τ -phase (Fe,Ni) ₅ Si ₂	τ -phase (Fe,Ni) ₅ Si ₂	(Fe,Ni) ₅ Si ₂	Ni ₅ Si ₂
	(Fe,Ni) ₂ Si	(Fe,Ni) ₂ Si	(Fe,Ni) ₂ Si	Ni ₂ Si
SiC	SiC	SiC	SiC	SiC

} + carbon

Table 1: Identification of the different phases formed within the product scale during the reaction between silicon carbide and iron-nickel alloys

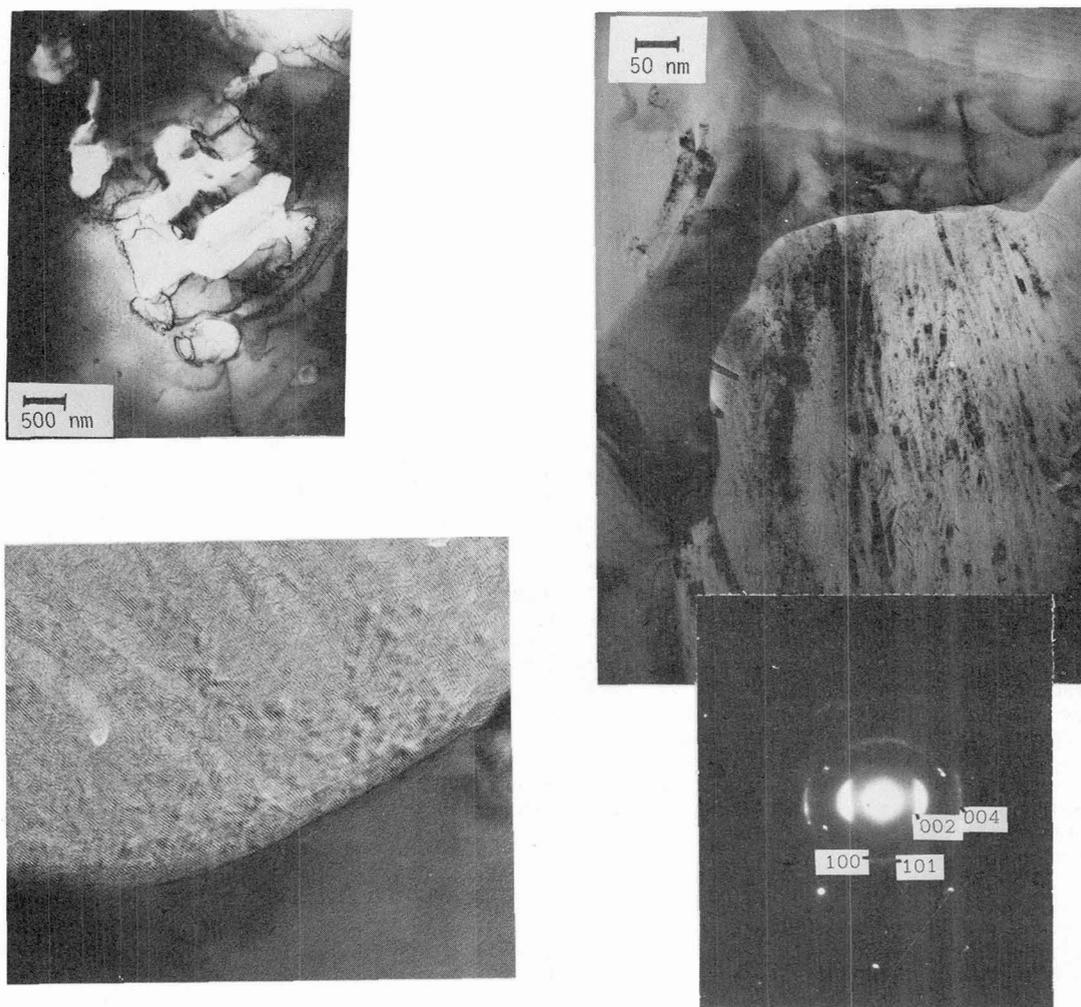


Fig.4 - Bright field image of a carbon precipitate in a τ -matrix (reaction couple Fe_{.75}Ni_{.25}-SiC, 1223 K, 4 h) with corresponding diffraction pattern and high resolution image.

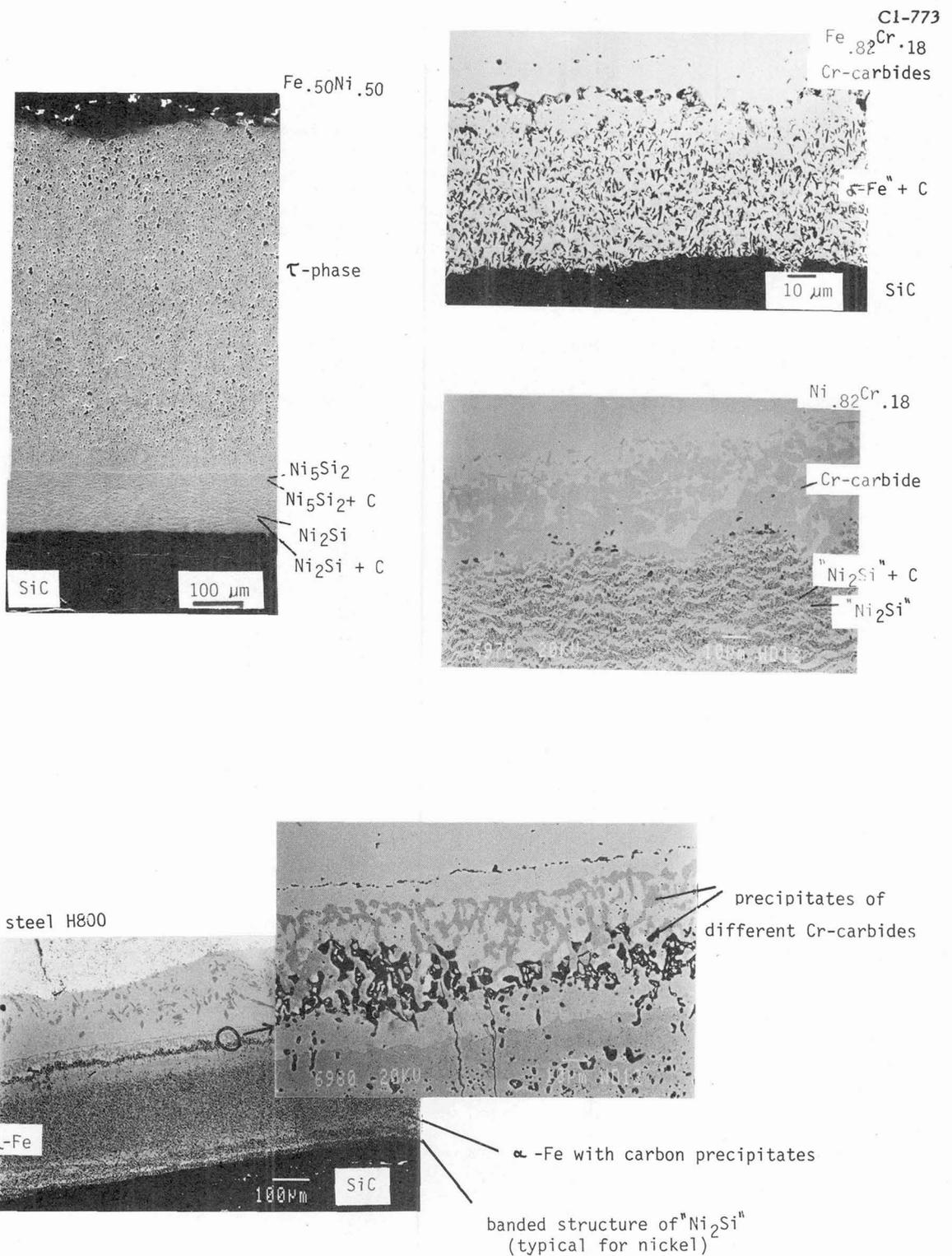


Fig.5 - Backscattered scanning electron micrograph of the reaction layer for the reactions of a) $(Fe_{.50}Ni_{.50}) - SiC$, b) $(Fe_{.82}Cr_{.18}) - SiC$, c) $(Ni_{.82}Cr_{.18}) - SiC$ and d) steel H800-SiC.

3 - CONCLUSION

The morphology features of the product scale for reactions between SiC and (Fe,Ni,Cr) alloys can be described as resulting of the present metal-components. Iron yields a broad region of α -region or τ -phase with randomly distributed carbon precipitates on the metal side of the reaction layer. The presence of nickel results in a banded zone near the SiC interface, bands of nickel-rich silicide alternate with carbon precipitate rich bands of the same silicide. Chromium finally precipitates at the border of the metallic interdiffusion zone in form of its various carbides.

REFERENCES

- (/1/ MEHAN, R.L., JACKSON, M.R., Mat. Sci. Res. 14 (1980) 513)
- (/2/ BENNETT, M.J., HOULTON, M.R. J. Mat. Sci. 14 (1979) 184)
- (/3/ MEHAN, R.L., MCKEE, D.W., J. Mat. Sci. 11 (1976) 1009)
- (/4/ SCHIEPER, R., VAN LOO, F.J.J., de WITJ, G., J. Am. Ceram. Soc. 71 (1988) C284)