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# Suessite, $Fe_3Si$: a new mineral in the North Haig ureilite

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## Abstract

The North Haig olivine pigeonite achondrite (ureilite) is a polymict breccia consisting of major olivine, low-Ca pyroxene and an intergranular carbonaceous matrix. Olivine and low-Ca pyroxene vary widely in composition, covering the ranges observed in all known ureilites. Minor granular enstatite clasts with diopside exsolution blebs, akin to enstatite achondrites, were also observed. Native metal in ureilites is normally kamacite of variable Ni content, in some cases with up to 2% Si in solid solution. However, kamacite with trace amounts of Si is extremely rare in North Haig, only a few 1–2 μm grains within silicates were observed. Instead, the common metallic phase is approximately $Fe_3Si$, a new mineral which we have named suessite in honor of Professor Hans E. Suess. Suessite occurs as minor anhedral vein fillings in interstitial cracks, in silicates, and in the intergranular carbonaceous material and ranges in size from 1 μm blebs to elongated grains about 30 × 150 μm in size. Suessite is cream white in reflected light, isotropic, ferromagnetic, and shows no cleavage. Reflectance in percent (determined by G. A. Desborough) at the 4 standard wavelengths of 470, 546, 589 and 650 nm for 2 grains is 48.5(5), 51.6(3), 53.5(7), 50(2), and 49.7(5), 53.4(4), 54.5(6), 52(1), respectively. Analyses (by EMX) indicate presence of dominating low-Ni and less common high-Ni varieties of suessite (in wt.%, mean in parentheses): Fe 84.7, 83.1 (84.2); Ni 1.6, 4.5 (2.5); Co 0.21, 0.27 (0.23); Cr 0.10, 0.04 (0.08); Si 15.3, 13.7 (14.7); P 0.06, 0.17 (0.10). Thus, suessite is $(\mathit{Fe}, \mathrm{Ni}, \mathrm{Co}, \mathrm{Cr})_{2.84-3.14}(\mathit{Si}, \mathrm{P})_{1.0}$, mean $(\mathit{Fe}, \mathrm{Ni}, \mathrm{Co}, \mathrm{Cr})_{2.96}(\mathit{Si}, \mathrm{P})_{1.0}$, very close to $Fe_3Si$. X-ray powder diffraction shows that suessite possesses a similar structure to alpha-Fe (kamacite) and the solid solution alloy $(Fe_3Si)_{ss}$ in displaying three lines (relative intensities in parentheses): 2.005(10), 1.42(1), 1.160(3) (in Å). The calculated cell size is 2.841±0.002Å ($V = 22.93Å^3$). Thus, suessite is the Si-rich end member of the α solid solution region of the Fe–Si phase diagram with composition close to $Fe_3Si$. Silicates, particularly olivine, in ureilites have core compositions of $Fo_{76-92}$ and thin (<100 μm) rims of essentially $Fo_{100}$ formed by reduction and reaction with the carbonaceous matrix material. We suggest that Si and Fe liberated in this reduction process formed suessite, possibly also by reaction with preexisting kamacite.

## Introduction

The North Haig olivine–pigeonite achondrite (ureilite), a meteorite found in 1961 in Western Australia (McCall and Cleverly, 1968), is a brecciated rock that consists of olivine and low-Ca pyroxene (pigeonite), embedded in an intergranular carbonaceous matrix. Olivine and pigeonite vary in composition over wide ranges and, in fact, cover the compositional ranges of all other ureilites combined (Berkley *et al.*, 1978a,b, 1980). North Haig also contains minor fragments of granular enstatite with diopside exsolution blebs, akin to some enstatite achondrites. Thus, North Haig consists of material of essentially all varieties of ureilites, in addition to minor amounts of clasts of non-ureilite origin, and is therefore properly classified as a polymict-brecciated ureilite.

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All ureilites contain a metal phase, kamacite, that occurs intimately associated with the intergranular carbonaceous matrix or as decorations along fractures and cleavage planes within silicate grains. This kamacite has variable Ni contents and up to about 3.8 wt.% Si in solid solution (Berkley *et al.*, 1980). However, electron microprobe analyses of the metallic phase in North Haig indicate that kamacite with trace amounts of Si in solid solution is extremely rare, and only a few 1–2 μm diameter grains were observed enclosed within silicate grains. Rather, the metallic phase in North Haig is $Fe_3Si$, a new mineral that also contains minor amounts of Ni(2.5), Co(0.23), Cr(0.08), and P(0.10; all avg. wt.%). In the present paper, we describe the properties of this new phase and discuss its origin. We have named this new mineral suessite, in honor of Dr. Hans E. Suess, Professor of Chemistry, University of California San Diego, La Jolla, California, USA, in recognition of his outstanding contributions to the field of cosmochemistry and meteoritics (Keil *et al.*, 1980). The mineral and name have been approved by the Commission on New Minerals and Mineral Names of the International Mineralogical Association.

## Occurrence and physical properties

Suessite generally occurs as anhedral vein fillings in interstitial cracks and fissures between silicate grains (Fig. 1). Most commonly it is found in the carbonaceous matrix, interstitial to silicate grains, rather than within them. Suessite ranges in grain size from 1 μm blebs to linear vein fillings (Fig. 1), occasionally up to several hundred μm in length. Other opaques in North Haig are rare troilite which occurs in blebs about 1–50 μm in diameter, both interstitially to and within silicate grains, as well as extremely rare 1–2 μm diameter blebs of low-Ni kamacite. Kamacite has less than 1 wt.% Ni and only trace amounts of Si (Table 1) and occurs exclusively within silicate grains, not in the interstitial carbonaceous matrix. In our sections, suessite was not observed in direct contact with either troilite or kamacite. North Haig is a meteorite find and, thus, has been subjected to considerable terrestrial weathering. Since metallic phases tend to most readily weather to hydrous ferric oxide, it is difficult to estimate the pre-terrestrial abundance of suessite and kamacite. However, in our sections, suessite is by far the most abundant metallic phase, whereas kamacite is extremely rare.

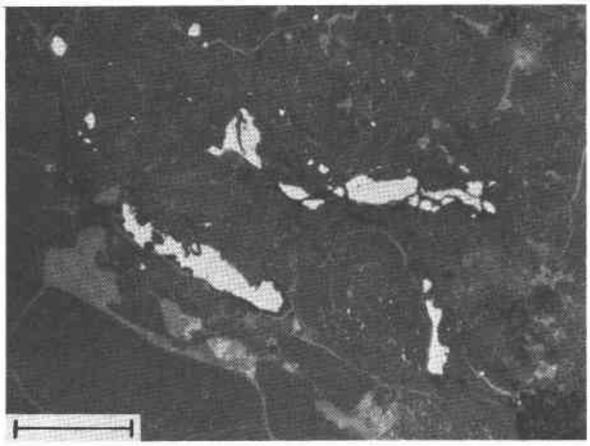

Fig. 1. Suessite (white) as vein fillings and blebs in the North Haig ureilite. Silicates (olivine, pigeonite) are gray; carbonaceous matrix is black. Hydrous ferric oxide veins (light gray) are of terrestrial weathering origin. Reflected, plane polarized light. Scale bar equals 100 μm.

In polished section, reflected light and air, suessite displays a cream white color under the microscope and is isotropic. Reflectance data for suessite for the four standard wavelengths were determined by G. A. Desborough, using the Zeiss standard WC-005. For two grains, the reflectance of suessite is as follows (in %): grain 1: 470–48.5(5), 546–51.6(3), 589–53.5(7), 650–50(2); grain 2: 470–49.7(5), 546–53.4(4), 589–54.5(6), 650–52(1) (numbers in parentheses are the number of measurements). Suessite shows slightly less relief under the microscope than associated silicate grains. The mineral does not display a discernable cleavage and is strongly ferromagnetic, *i.e.*, is significantly affected by the magnetic field of a small hand magnet.

## Composition

The composition of suessite was determined with an automated ARL EMX-SM electron microprobe, using an accelerating potential of 15 kV and a sample current of about 0.03 μA. Standards were pure Fe, Si, Ni, Co, and Ca, and P in natural apatite. Synthetic $Fe_3Si$, prepared by annealing under pressure of stoichiometric amounts of pure Fe and Si, was employed as a secondary standard. Results obtained by analyzing suessite against pure Fe and Si or the synthetic $Fe_3Si$ compared very well, after appropriate corrections for differential matrix effects were made. Corrections for instrumental and differential matrix effects were made following the procedures described by Keil (1967), using the Tracor Northern ZAF correction procedure for differential matrix effects.

Table 1. Electron microprobe analyses of suessite, minor troilite and extremely rare kamacite in the North Haig ureilite (in wt.%). Numbers in brackets are numbers of grains analyzed.

| | Suessite | | | Troilite | Kamacite |
|---|---|---|---|---|---|
| | Low Ni | High Ni | Mean | | |
| Fe | 84.7 (83.0 -88.8 ) [47] | 83.1 (81.7 -85.8 ) [20] | 84.2 [67] | 60.8 | 98.5 |
| Ni | 1.6 ( 0.5 - 2.4 ) [47] | 4.5 ( 2.9 - 6.4 ) [20] | 2.5 [67] | < 0.06 | 0.51 |
| Co | 0.21( 0.16- 0.30) [32] | 0.27( 0.20- 0.32) [11] | 0.23[43] | 0.10 | 0.24 |
| Cr | 0.10(< 0.02- 0.25) [30] | 0.04( 0.02- 0.06) [ 8] | 0.08[38] | 0.12 | 0.08 |
| Si | 15.3 (14.6 -16.4 ) [26] | 13.7 (12.6 -14.3 ) [15] | 14.7 [41] | n.d. | 0.07 |
| P | 0.06(< 0.02- 0.12) [26] | 0.17( 0.10- 0.25) [15] | 0.10[41] | n.d. | n.d. |
| S | n.d. | n.d. | n.d. | 37.5 | n.d. |
| TOTAL | 101.97 | 101.78 | 101.81 | 98.52 | 99.40 |
| | | Atomic ratios of (Fe, Ni, Co, Cr) : (Si,P) | | | |
| | 2.84 : 1 | 3.14 : 1 | 2.96 : 1 | | |

Composition of suessite is shown in Table 1 and Figures 2–4. Suessite contains major Fe, Ni and Si and minor to trace amounts of Co, Cr and P. Although compositions vary from grain to grain, analyses indicate existence of two varieties of suessite, one with low Ni (avg. 1.6 wt.%), low Co and P, and high Fe, Cr and Si; and one with high Ni (avg. 4.5%), high Co and P, and low Fe, Cr and Si. Random analyses indicate that low-Ni suessite is about 2.5 times as abundant in our sections of North Haig as is high-Ni suessite. The compositional gap between low- and high-Ni varieties is at about 2.7 wt.% Ni (*e.g.*, Fig. 2), but both low- and high-Ni suessite vary slightly in composition from grain to grain, covering a range of 0.5–6.4 wt.% Ni (Table 1; Figs. 2–4). Nickel apparently substitutes for Fe, but the correlation is far from perfect (r = 0.56; Fig. 2). Cobalt correlates well with Ni (r = 0.69; Fig. 3) and increases with increasing Ni (and decreases with increasing Fe), and P substitutes for Si (r = 0.85; Fig. 4). Chromium is slightly higher in low-Ni than in high-Ni suessite (avg. 0.10 *vs.* 0.04 wt.%, respectively), but precision of routine microprobe analy-

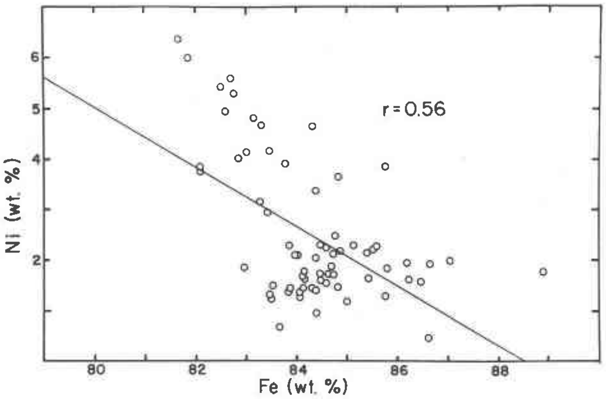

Fig. 2. Nickel and iron contents of 47 grains of low-Ni and 20 grains of high-Ni suessite from the North Haig ureilite, as determined by electron microprobe analysis. Boundary between low- and high-Ni suessite is at ~2.7% Ni. Diagonal line is a multivariate best-fit regression line; r = statistical correlation coefficient.

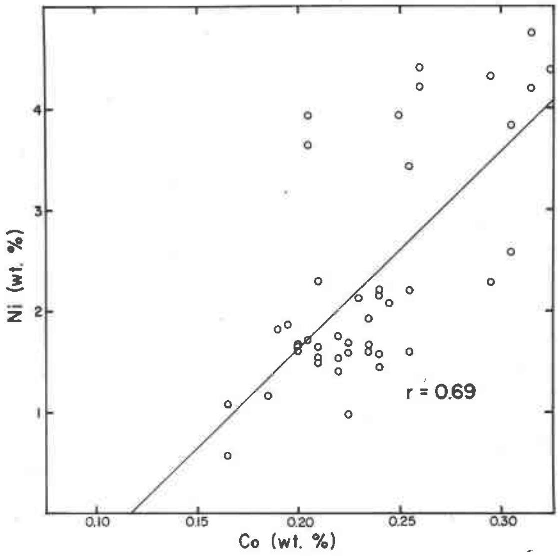

Fig. 3. Nickel and cobalt contents of 32 grains of low-Ni and 11 grains of high-Ni suessite from the North Haig ureilite as determined by electron microprobe analysis. Symbols as in Fig. 2.

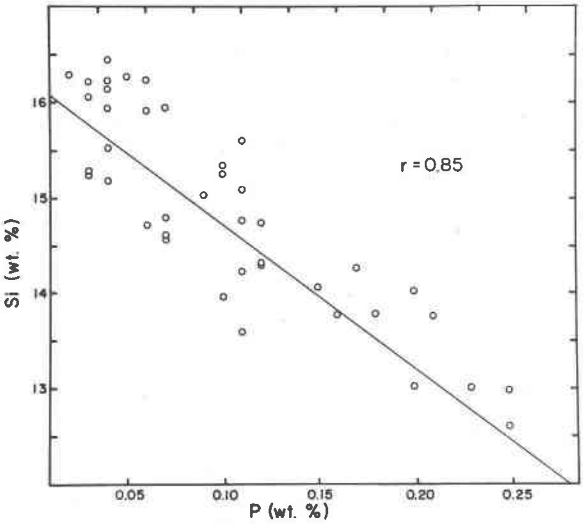

Fig. 4. Silicon and phosphorous contents of 26 grains of low-Ni and 15 grains of high-Ni suessite from the North Haig ureilite as determined by electron microprobe analysis. Symbols as in Fig. 2.

ses at these low concentrations is too low to reveal any systematic compositional correlations. In the highly reduced environment in which suessite formed, it appears likely that chromium occurs in the divalent rather than the trivalent state. The atomic ratios for suessite were therefore calculated on the basis of (Fe, Ni, Co, Cr):(Si, P), with the atomic proportions of Si + P = 1. These ratios are 2.84:1 for low-Ni suessite, 3.14:1 for high-Ni suessite, and 2.96:1 for the weighted mean. The structural formulae are as follows:

Low-Ni suessite
$(Fe_{2.778}Ni_{0.049}Co_{0.007}Cr_{0.004})(Si,P)_{1.0}$
High-Ni suessite
$(Fe_{2.976}Ni_{0.154}Co_{0.01}Cr_{0.002})(Si,P)_{1.0}$
Mean suessite
$(Fe_{2.867}Ni_{0.082}Co_{0.008}Cr_{0.004})(Si,P)_{1.0}$

Simplified, mean suessite is therefore $(Fe_{2.9}Ni_{0.1})$ Si or nearly $Fe_3Si$. Thus, suessite is compositionally identical to the Si-rich end member of the α solid solution region of the Fe–Si phase diagram, namely the compound close to $Fe_3Si$, with minor substitutions of Ni and trace substitutions of Co, Cr, and P. That Si-rich boundary in the Fe–Si system, according to Hansen (1958), is near 26 atomic percent Si, very near the 25.1 atomic percent Si calculated for mean suessite.

## Structure

X-ray powder diffraction studies of suessite were performed by scratching small amounts of powder from the interiors of several of the largest analyzed grains. The powder was picked up on a grease-coated glass fiber mount. The X-ray powder pattern (Fe*K*α radiation) was obtained using diamond powder as an internal standard for determining *d*-spacings (Table 2).

The resulting X-ray patterns show that suessite possesses a similar structure to alpha-Fe (kamacite) and to the solid solution alloy $(Fe_3Si)_{ss}$ in displaying only three lines as compared to six for the intermetallic stoichiometric compound $Fe_3Si$ ($BiF_3$-type; b.c.c., p. 712, Hansen, 1958) (Table 2). The calculated cell size for suessite is 2.841±0.002Å ($V$ = 22.93Å$^3$) which corresponds to 18–20 atomic percent of Si in pure Fe (Pearson, 1958), in approximate agreement with the analyzed grains (Table 1). The combined effect of impurities of Ni, Co, Cr and P on *d* values is not known, but this may cause the discrepancy in values for atomic percent Si predicted on the basis of the *d* value and measured by electron microprobe.

Our X-ray diffraction data seem to suggest that suessite is not the intermetallic compound $Fe_3Si$ ($BiF_3$-type), although it very closely approaches this composition, but rather a solid solution in the system Fe–Si. However, the X-ray data are not unambiguous. The 3 missing lines of the intermetallic compound in the suessite pattern are very weak and it is doubtful that we would have seen them even if they occurred, because of the high background in the suessite pattern (note that we were

Table 2. X-ray powder diffraction data for suessite in the North Haig meteorite compared with the intermetallic compound $Fe_3Si$, and $Fe_3Si$ solid solution alloy and α-Fe.

| Suessite<br>N. Haig<br>d Å | (1)<br>I | $Fe_3Si$<br>$BiF_3$-type<br>d Å | (2)<br>I | $(Fe_3Si)_{ss}$<br>d Å | (3)<br>I | α-Fe<br>d Å | (4)<br>I |
|---|---|---|---|---|---|---|---|
| -- | -- | 3.26 | 0.6 | -- | -- | -- | -- |
| -- | -- | 2.82 | 0.3 | -- | -- | -- | -- |
| 2.005 | 10 | 1.994 | 10 | 1.998 | 10 | 2.027 | 10 |
| -- | -- | 1.70 | 0.3 | -- | -- | -- | -- |
| 1.42 | 1 | 1.41 | 1.5 | 1.413 | 1.5 | 1.433 | 1.5 |
| 1.160 | 3 | 1.151 | 3 | 1.154 | 3 | 1.170 | 3 |

(1) Norelco powder camera, diam. 57.3 mm, Fe K radiation, Mn filter; Diamond internal standard; intensities (I) are visual. a = 2.841 ± 0.002 Å ( α - Fe = 2.886 Å). (2) - (4) Calculated patterns by B. Tani.

able to separate only minute amounts of the phase from the meteorite and, hence, long exposure times were necessary). Furthermore, the synthetic $Fe_3Si$ prepared by hot pressing of pure Fe and Si in the stoichiometric proportions also gives an X-ray pattern identical to that of suessite and with the 3 weakest lines of the intermetallic compound missing. Thus, the possibility exists that the particular annealing history accounts for the absence of these lines in the synthetic and natural phase. Finally, according to Hansen's (1958) review of the Si-rich end member of the α solid solution region of the Fe–Si phase diagram, there is a controversy as to whether or not the disorder–order transition occurs within a homogeneous phase of the simple bcc (α) type. However, there is agreement that the Si-rich boundary of this region is at 26 atomic percent Si near $Fe_3Si$. We therefore define suessite as that phase.

## Discussion

Silicides are stable under exceedingly low oxygen partial pressures and, thus, are not likely to form in crustal rocks of the earth. Nevertheless, several terrestrial occurrences of silicides have been reported. The first study describes FeSi and $FeSi_2$ from placers and drill core samples of sandstones of the Poltava series, Donets Region (Gevork'yan, 1969; Gevork'yan *et al.*, 1969), but these occurrences have been questioned by Fleischer (1970, 1980). Additional occurrences of FeSi as well as of $Fe_4Si_9$, $Fe_2Si$ and $Fe_3Si_2$ were more recently described from acid insoluble residues of Lower Cambrian red limy sandstones and limestones from the Bazaikha River, western part of eastern Sayan (Novoseleva, 1975; Novoseleva and Bagdasarov, 1979) and were attributed by these authors to an extraterrestrial origin due to ablation of extraterrestrial silicides during passage through the earth's atmosphere. However, this appears to be an unlikely mode of origin, in view of the exceedingly low abundance of silicides in meteorites.

Only one silicide has previously been described from meteorites, namely the mineral perryite, a nickel silicide containing minor iron substituting for Ni, and P for Si. The mineral was found exclusively as a very rare compound in highly-reduced meteorites such as irons, pallasites, enstatite chondrites and enstatite achondrites (*e.g.*, Fredriksson and Henderson, 1965; Reed, 1978; Wai, 1970). The accepted composition, originally given as $(Ni,Fe)_2(Si,P)$ (Fredriksson and Henderson, 1965; Reed, 1968) is now considered to be $(Ni,Fe)_5(Si,P)_2$ (Wai, 1970).

The new mineral suessite, $Fe_3Si$, also formed under highly reducing conditions. Silicates in ureilites, particularly olivine, have thin (<100 μm) rims in contact with the carbonaceous matrix that are essentially FeO-free, whereas the main portions of the grains have moderately high FeO contents (*e.g.*, Berkley *et al.*, 1976, 1978a,b, 1980). Specifically, olivine rim compositions are essentially $Fo_{100}$, whereas the remainder of the grains is about $Fo_{76-92}$. We and others have suggested that the FeO-free rims formed by reduction of iron by the carbonaceous matrix. We propose that suessite also formed in this process, due to reduction of iron and stoichiometrically equivalent silicon liberated in the reduction process. Possibly, reaction of liberated iron and silicon with preexisting kamacite may also have taken place in the formation of suessite. Hence, only the few minute grains of kamacite occurring *within* silicate grains were sufficiently shielded to remain as kamacite and did not react to form suessite.

It is curious, however, that the polymict-brecciated ureilite North Haig is the only one that contains suessite, whereas the other seven studied by us (Berkley *et al.*, 1980) contain kamacite with <0.03–3.8 wt.% Si. Special conditions may have prevailed during the formation of the North Haig breccia that did not occur or only occurred to a lesser degree during the formation of the other ureilites. All ureilites show evidence for shock metamorphism, but North Haig is by far the most severely shocked ureilite. The formation of suessite may have occurred during this extreme shock event which may have caused rapid increase in temperature with concomitant reduction of silicate rims by carbon from the matrix, followed by rapidly falling temperatures. The varying composition of suessite, the occurrence of only thin FeO-free rims around silicates, and the presence of some remnant kamacite suggest that this process was indeed of rather limited duration and was probably followed by a rapid drop in temperature.

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