Euhedral tetrataenite in the Jelica meteorite

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Abstract

A 65 × 107 μm grain of euhedral tetrataenite (ordered FeNi) attached to a similarly sized grain of troilite occurs within an impact-melt rock clast in the Jelica LL6 chondrite breccia. After impact melting, immiscible metallic Fe-Ni and troilite droplets formed within the silicate melt progenitor of the clast. At $\geqslant$1200°C, while the surrounding silicate was still partly molten, euhedral taenite with ~50 wt.% Ni began crystallizing in one of the metal–troilite droplets. Troilite nucleated at one edge of the euhedral taenite grain and began to crystallize at ~870°C. At 320°C the metal phase underwent an ordering reaction and formed tetrataenite. The unrecrystallized clast–host boundary and the differences in olivine composition and degree of polycrystallinity of troilite between the clast and Jelica host indicate that the clast was incorporated into Jelica during a late-stage brecciation event.

KEYWORDS: tetrataenite, troilite, Jelica meteorite, brecciation.

Introduction

MOST metallic Fe-Ni grains in solar system rocks are anhedral. In terrestrial rocks metallic Fe-Ni itself is rare, being largely restricted to serpentinized peridotites (e.g. Krishnarao, 1964; Frost, 1985) and volcanic rocks that assimilated carbonaceous material (e.g. Goodrich and Bird, 1985). Although metallic Fe-Ni is ubiquitous among lunar rocks, its modal abundance in essentially every sample is < 1 vol.% (Heiken *et al.*, 1991). In lunar basalts and eucrites (meteoritic basalts), most of the metallic Fe-Ni occurs interstitially between silicate grains and thus tends to have irregular morphologies (Duke, 1965). In many porphyritic chondrules, metallic Fe-Ni and troilite form rounded blebs in the mesostasis because their precursors were immiscible droplets (e.g. Gooding, 1979). In metamorphosed ordinary chondrites, metallic Fe-Ni and troilite typically form anhedral grains. Some of the metallic Fe-Ni and troilite in meteorites and lunar rocks has also been mobilized and injected into fractures in adjacent silicate grains where local shock-reheating temperatures reached the Fe–FeS eutectic (988°C). In interplanetary dust particles, metallic Fe-Ni occurs most commonly along with sulphide as spheroids and fragments.

Euhedral metallic Fe-Ni grains are thus relatively rare. This is because several restrictive conditions must be met before such grains can form. Because of these restrictions, the occurrence of euhedral metallic Fe-Ni grains in an object can potentially provide important petrogenetic information.

(1) *Grain growth must occur at free surfaces*, generally restricting euhedral grains to systems that are igneous or undergoing vapour deposition. There are two possible exceptions to this generalization. Nagata *et al.* (1991) demonstrated that tiny (~0.015 μm) cubes of tetrataenite could be synthesized by coalescence growth from a joint cloud of smoke-size particles of Fe and Ni generated by evaporation; they suggested that analogous processes could have occurred in the solar nebula, but the generation of independent clouds of pure Fe and Ni particles seems highly unlikely. Although idioblastic metallic Fe-Ni grains could conceivably have formed in metamorphosed chondrites, such grains have not been reported. Nevertheless, Christophe Michel-Lévy (1979, 1981) reported subhedral metallic Fe-Ni grains in the matrices of a few relatively unshocked H4 and H5 chondrites; these grains display crystal faces on edges which protrude into voids in the hosts.

(2) *The metal ± sulphide assemblage must have an appropriate bulk composition* so that taenite is the liquidus phase in igneous systems or the stable condensate phase in vapour-deposition systems.

Mineralogical Magazine, June 1994, Vol. 58, pp. 215–221

(3) *Metallic Fe-Ni grains must remain undeformed* during subsequent compaction, thermal metamorphism, aqueous alteration and shock.

Despite its rarity, euhedral metallic Fe-Ni occurs in a wide variety of extraterrestrial materials. Some of these (i.e. grains within chondrules) formed in the solar nebula; others (e.g. grains in vugs, lunar glass spherules, deep-sea spherules and impact melt-rock clasts) formed on parent body surfaces by meteoroid impacts. Housley (1981) reported curvilinear trails of tiny (2 μm) crystals of awaruite (Ni_3Fe) within a fracture in an olivine grain from a porphyritic olivine chondrule in the Allende CV3 chondrite. Within another Allende porphyritic olivine chondrule, Rubin (1991) found a large (290 × 510 μm) magnetite- and pentlandite-rich nodule containing 9 vol.% euhedral awaruite grains typically 35–65 μm in size. Euhedral low-Ni kamacite grains with trapezohedral, cubic, tetrahexahedral, octahedral and dodecahedral faces occur within vugs in highly recrystallized Apollo 14, 15 and 16 lunar breccias where they formed by vapour deposition of Fe onto silicate grain surfaces (Clanton *et al.*, 1973). Analogously, vugs within the Farmington L5 fragmental breccia contain elongated prismatic and (possibly) trapezohedral kamacite grains attached to a silicate substrate (Olsen, 1981). Ivanov (1989) reported several 350 μm size euhedral metallic Fe-Ni grains with martensitic compositions inside a 2 mm diameter vug located within a fracture in the CR2-like lithology of the Kaidun breccia (i.e. Kaidun I). Glass (1971) reported the occurrence of about 25 ~4 μm size octahedral crystals of kamacite situated along a plane within a 230 μm diameter pale green glass spherule from Apollo 11 sample 10084. He also found similar octahedral metallic Fe-Ni crystals in two glass particles from sample 12057, one of which contains hundreds of tiny (2 μm) crystals occurring along a nearly hemispherical surface. Frondel *et al.* (1970) reported 5 μm size octahedral crystals of metallic Fe in a glass fragment from Apollo 11 fines; isolated subhedral metallic Fe crystals reported in these fines may have been detached from glass surfaces. Glass (1982) reported 1–2 μm octahedral metallic Fe-Ni grains in Late Eocene clinopyroxene-bearing spherules from the equatorial Pacific Ocean; the number of octahedral metal grains in individual spherules ranges from a few to more than 100.

Here I report the first known natural occurrence of euhedral tetrataenite (ordered FeNi).* Tetrataenite is a common phase in chondrites and mesosiderites (brecciated stony-iron meteorites) where it occurs as irregular grains in contact with kamacite, taenite, troilite and/or silicate; it also occurs as rims on taenite grains in chondrites, diogenites, mesosiderites, pallasites and irons (Clarke and Scott, 1980).

Analytical procedures

For the purpose of an unrelated investigation, a 2 × 2.5 cm polished slab of Jelica was examined in reflected light, revealing the presence of a euhedral grain of tetrataenite. Polished thin-section UCLA 360 of Jelica was also examined microscopically. The slab was etched with 2% nital for examination of metal structures. After the slab was coated with C, mineral phases were analysed with the UCLA Cameca Camebax-microbeam electron microprobe using crystal spectrometers, 20-s counting times, ZAF corrections and a sample current ranging from 10 to 15 nA at 20 kV. Silicate phases were analysed with the following natural standards: magnetite (Fe), forsterite (Mg), grossular (Ca,Al,Si), jadeite (Na) and orthoclase (K). Plagioclase was analysed with a 5 μm beam; all other phases were analysed with a focused beam. Tetrataenite and troilite were analysed with the following standards: pure Fe, pure Ni, pure Cu, NBS steel 1156 for Co (7.3 wt.%) and Canyon Diablo troilite for S (36.5 wt.%). Cobalt background counts were taken at wavelengths of 177.4 and 180.8 μm; these wavelengths were found to be the most suitable for the elimination of the contribution of the Fe-K_β peak to the Co-K_α peak. Backscattered electron images were also made with the Cameca probe.

Results

Petrography of Jelica

Jelica is extensively recrystallized, consistent with its petrologic type-6 classification (Graham *et al.*, 1985). Nevertheless, some 400–2000 μm diameter porphyritic olivine (types I and II), porphyritic olivine-pyroxene, barred olivine and radial

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* After this paper was submitted, M.E. Zolensky (pers. comm., 1993) informed me that he found an unusual chondritic interplanetary dust particle (IDP) containing a 0.05 μm cube of metallic Fe–Ni with 58 wt.% Ni (probably tetrataenite) surrounded by pyroxene- and feldspar-normative glass. It is unclear how this IDP formed; it could be a nebular condensate (possibly derived from a comet) or a fragment of an impact melt-rock from an oxidized chondritic asteroid.

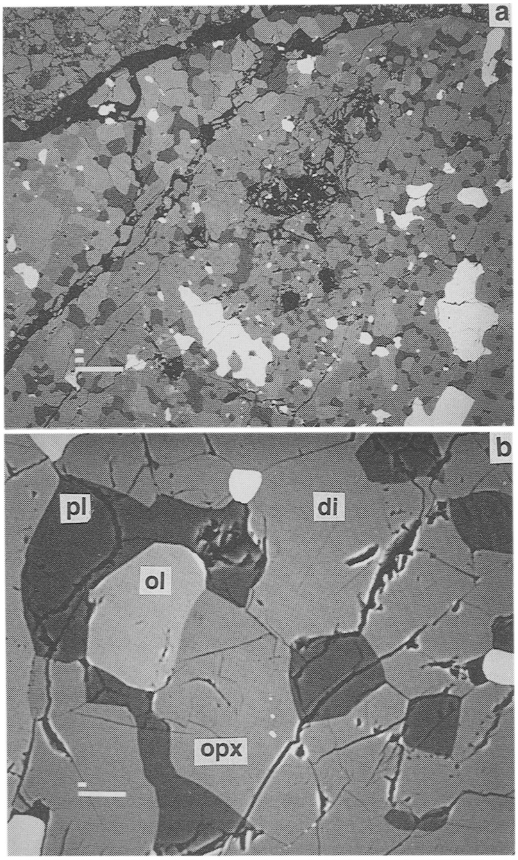

FIG. 1. Backscattered electron images of clast A in Jelica. (*a*) Low-magnification image showing the hypidiomorphic-granular texture. Metallic Fe-Ni and troilite (white), plagioclase (dark gray), orthopyroxene and diopside (medium gray) and olivine (light gray) are present. The boundary between the igneous-textured clast and the clastic breccia host (upper left) is sharp but marked by weathering veins (black) which also penetrate the clast. The euhedral tetrataenite grain and attached troilite are at lower right. Long white scale bar at lower left is 100 μm in length. (*b*) Higher-magnification image of part of the lower right region of Fig. 1*a* illustrating the nonclastic texture. Long white scale bar at lower left is 10 μm in length. pl = plagioclase; ol = olivine; opx = orthopyroxene; di = diopside. White grains are metallic Fe-Ni.

pyroxene chondrules and chondrule fragments are still discernible. Jelica is also brecciated; angular chondritic clasts (200–6500 μm), chondrules, isolated mineral grains and grain fragments are embedded in a clastic matrix composed of fragmented 10–20 μm size grains of olivine, pyroxene and plagioclase. Metallic Fe-Ni, troilite and chromite occur in both the matrix and the chondritic clasts. One 10 × 20 μm grain of metallic Cu was observed at a boundary between metallic Fe-Ni and troilite.

It is clear that Jelica has been shocked. Many troilite grains are polycrystalline and some exhibit deformation twins under partially crossed polars. Olivine grains exhibit undulose extinction; some also contain planar fractures indicating that Jelica should be classified as shock stage S3 (Stöffler *et al.*, 1991); this corresponds to a shock pressure of 15±5 GPa and a minimum temperature increase of 60±40°C. Opaque veins, which occur in the chondritic clasts, include thin (1–2 μm) blebby veins of troilite up to 180 μm long and rare veins of chromite, up to 80 μm long, within olivine grains. The presence of these opaque veins indicates that localized temperature concentrations during shock-heating must have been much higher than the minimum, i.e. at least 988°C and 1645°C for veins of metallic Fe-Ni-troilite and chromite, respectively. One 100 × 350 μm chondritic clast is almost completely opaque due to extensive silicate darkening. Numerous 0.2 × 10 μm veins of troilite and rare intergrown metallic Fe-Ni occur within silicate grains and along grain boundaries, and in rare cases the opaque veins cut across adjacent silicate grains.

The high mean fayalite (Fa) content of olivine (32.4±0.3 mol.%) and the absence of normal kamacite (two low-Ni metallic grains with 9.6 and 30.0 wt.% Co have been reported; Sears and Axon, 1976; Rubin, 1990) are consistent with Jelica's LL classification and indicate that this meteorite is one of the most oxidized ordinary chondrites. The bulk chemical composition (Kallemeyn *et al.*, 1989) is also consistent with an LL classification.

Characteristics of clast with euhedral tetrataenite

Clast A is a fractured, nearly triangular object in plane view, approximately 3.9 × 6.0 mm in size. Its boundary with surrounding mafic silicate mineral fragments in the Jelica host is sharp and unrecrystallized. The clast consists principally of olivine, orthopyroxene, diopside and plagioclase, forming a hypidiomorphic-granular texture (Fig. 1). No relict chondrules are present. Opaque phases, which include metallic Fe-Ni, troilite and chromite, constitute ~2 vol.% of the clast; most grains occur interstitially to the silicates, and, with one exception, all have irregular shapes. Unlike

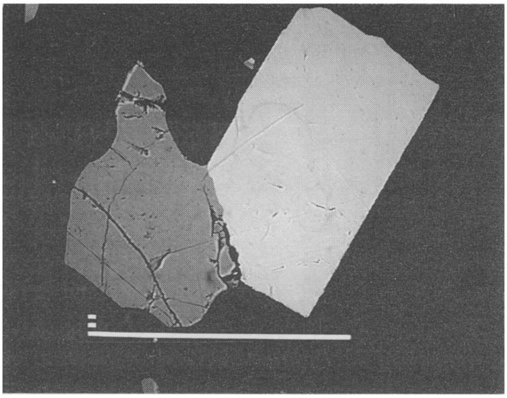

FIG. 2. Backscattered electron image of the euhedral tetrataenite grain (white) and attached troilite (medium gray). Surrounding silicate is black. The tetrataenite grain has the morphology of a truncated rectangle in plan view; the troilite nucleated and grew at the truncated side of the tetrataenite grain. Long white scale bar at lower left is 100 μm in length.

the troilite grains in the Jelica host, those in clast A are monocrystalline and do not exhibit deformation twins.

Although clast A contains several grains of tetrataenite, only one is euhedral (Fig. 2). This 65 × 107 μm size monocrystalline grain has the shape (in plane view) of a truncated rectangle. At the truncated edge, the tetrataenite grain is attached to an irregularly shaped 65 × 105 μm size monocrystalline grain of troilite. The tetrataenite grain abuts surrounding grains of plagioclase, olivine and pyroxene. The tetrataenite–troilite assemblage consists of 61 vol.% (73 wt.%) tetrataenite and 39 vol.% (27 wt.%) troilite. (Calculated mineral densities of 8.275 and 4.83 g cm^{-3}, respectively, were assumed; Nickel and Nichols, 1991).

Mineral chemistry and bulk composition

The euhedral tetrataenite grain contains 2.0 wt.% Co and 0.13 wt.% Cu (Table 1). Such a high Co content is not unusual for Ni-rich metal in highly oxidized chondrites (Sears and Axon, 1976; Rubin, 1990, 1991). The Cu content is near the lower extreme of Cu values reported for meteoritic tetrataenite (0.11–0.36 wt.%; Table 1 of Clarke and Scott, 1980). The euhedral grain is zoned in Fe: grain edges average 47.6 wt.% Fe compared to 49.7 wt.% Fe at grain centres; thus, there is a 4.2% relative depletion of Fe at grain edges. Other grains of (anhedral) tetrataenite both within and outside of clast A have similar compositions (Table 1). Troilite, the other major opaque phase in the clast, is essentially stoichiometric FeS.

Olivine in clast A (Fa_{31}) is somewhat more magnesian than olivine in the Jelica host (Fa_{32}). Plagioclase in clast A is compositionally zoned, e.g. the core of one large grain ($An_{12.2}$) is 18% relatively more anorthitic than the grain edge ($An_{10.0}$).

The bulk composition of the tetrataenite–troilite assemblage was calculated from the phase compositions (Table 1) and the modal abundances (in wt.%; see above); this value (Table 2) is somewhat uncertain because of the lack of information on the assemblage's three-dimensional structure.

Discussion

Formation of euhedral tetrataenite

The most likely mechanism for forming the euhedral tetrataenite grain is by primary crystallization from a melt. This requires that the metallic Fe-Ni-sulphide assemblage in clast A formed igneously. The clast's hypidiomorphic-granular texture and lack of chondrules is consistent with formation from a melt. Because Jelica is shocked,

TABLE 1. Mean compositions (wt.%) of tetrataenite in Jelica.

	Clast A		Outside clast A
	Euhedral	Others	
no. of points	10	2	4
Fe	49.1 ± 0.8	50.7	48.9
Co	2.0 ± 0.0	1.6	2.0
Ni	49.7 ± 0.9	48.3	50.0
Cu	0.13 ± 0.02	0.17	0.15
Total	100.9	100.8	101.0

brecciated and known to contain impact-melt rock clasts (Fodor and Keil, 1975), it is reasonable to conclude that clast A is also an impact-melt rock. Furthermore, some impact-melt-rock clasts in ordinary chondrites are analogous to clast A in containing olivine that is more magnesian than that in the host (e.g. Rubin, 1985). The most probable progenitor for clast A is the Jelica whole rock.

During cooling from high temperatures, the silicate portion of clast A underwent fractional crystallization. This is indicated by the normal compositional zoning of individual plagioclase grains, and, possibly, by the Fe enrichment at the edges of the euhedral tetrataenite grain.

Impact melts of chondritic material separate into immiscible silicate and metallic Fe-Ni–troilite liquids; the euhedral tetrataenite grain in clast A must have begun crystallizing within one such metallic-Fe-Ni- and troilite-rich droplet. The euhedral morphology of this grain suggests that the precursor droplet itself was not confined interstitially between solidified silicates. In order to demonstrate that the surrounding silicate was still partly molten, it must be shown that the liquidus of the opaque assemblage containing the euhedral tetrataenite grain is above the solidus of the Jelica host.

Melting experiments on the St. Séverin LL6 chondrite were undertaken by Jurewicz *et al.* (1993). The experiments were run at atmospheric pressure and the oxygen fugacity was fixed at 1 log unit below the iron–wüstite buffer. Although these experiments were not run at temperatures below 1170°C, there was definitely melt present in the charge at this temperature. Hence, the St. Séverin solidus must be below 1170°C.

Because Jelica is slightly more oxidized than St. Séverin (their mean olivine compositions are 32.4 and 30.3 mol.% Fa, respectively; Rubin, 1990), its solidus is probably somewhat lower. I estimate that the Jelica solidus is 1130–1150°C. Thus, impact-melt rocks that formed in the Jelica regolith (i.e. total melts with the same bulk composition as Jelica) should still contain some silicate liquid before reaching this temperature during cooling.

TABLE 2. Bulk composition (wt.%) of opaque assemblage consisting of troilite and euhedral tetrataenite.*

S	9.8
Fe	52.6
Co	1.5
Ni	36.0
Cu	0.1

* Determined from modal abundances, mineral compositions and mineral densities; normalized to 100%.

The bulk composition of the euhedral-tetrataenite-bearing assemblage (Table 2) can be plotted on the Fe–Ni–S phase diagrams of Kullerud *et al.* (1969). On the highest temperature diagram available (1100°C) (Fig. 14 of Kullerud *et al.*, 1969), the assemblage composition plots near the centre of the taenite + liquid field. Because this field is so large (it constitutes 40% of the Fe–Ni–S diagram at this temperature), it seems reasonable to infer that the field persists to temperatures significantly higher than 1100°C. This inference is consistent with the Fe–S and Ni–S binary phase diagrams (Figs. 1 and 4 of Kullerud *et al.*, 1969) which show that compositions with comparable amounts of S (i.e. ~10 wt.%) are in the metal + liquid stability fields at temperatures exceeding 1250°C. Although the lack of experimental data precludes concrete demonstration that the metallic Fe-Ni grain would have begun crystallizing while silicate liquid was still present, it nevertheless seems very probable that this was the case.

Within the cooling metallic-Fe-Ni–troilite droplet, metallic Fe-Ni with ~50 wt.% Ni began to crystallize as taenite (fcc); this probably started at a temperature ⩾1200°C. The large difference in surface energy between the crystallizing taenite and the surrounding increasingly S-rich liquid was responsible for the development of euhedral faces on the taenite grain. As the system cooled by several hundred degrees, taenite remained the only liquidus phase. Interpolation between the 900°C and ~860°C Fe–Ni–S phase diagrams (Figs. 67 and 68 of Kullerud, 1963) indicates that troilite began crystallizing from the melt when temperatures reached about 870°C. The absence of pentlandite in the assemblage probably indicates that this phase failed to crystallize at lower temperatures; it also suggests inaccuracies in the troilite–taenite, troilite–pentlandite–taenite and pentlandite–taenite tie lines as drawn by Kullerud (1963). Troilite nucleated at one edge of the taenite grain and probably incorporated all of the remaining sulphide-rich liquid surrounding the taenite grain. The 4% relative decrease in Fe at the edge of the euhedral metal grain relative to its centre could either be a result of igneous zoning due to fractional crystallization (as in the case of plagioclase in the clast) or of subsequent solid-state diffusion of Ni from nearby troilite grains during mild annealing.

When temperatures reached ~460°C the taenite transformed into the disordered γ′ FeNi phase (Reuter *et al.*, 1988); at 320°C the γ′ phase underwent an ordering reaction and formed tetrataenite (ordered FeNi).

Incorporation of clast A into the Jelica host

Because olivine equilibrates rapidly during metamorphism (e.g. Freer, 1981), the minor difference in olivine composition between clast A and the Jelica host indicates that the clast was not metamorphosed along with the rest of Jelica. This is consistent with the sharp boundary between the clast and host. The essential absence of polycrystallinity in troilites in clast A indicates that the clast also did not experience the same shock history as the host. It thus seems probable that the clast was incorporated into the Jelica host during a mild late-stage brecciation event. The clast is thus analogous to aberrant chondrules and metal and silicate grains in type 4–6 ordinary chondrites which have compositions significantly out of equilibrium with those in the host (e.g. Scott *et al.*, 1985; Rubin, 1990). Like clast A in Jelica, these objects must have been incorporated after their hosts cooled from high metamorphic temperatures.

Acknowledgements

I thank A. Treiman for once pointing out that R. Housley reported euhedral awaruite grains 10 years before I did. I am also grateful to J. T. Wasson, D. W. Mittlefehldt, P. H. Warren, A. N. Krot, B. P. Glass and M. E. Zolensky for helpful discussions and to M. Christophe Michel-Lévy for her correspondence. Reviews by R. Hutchison and an anonymous referee were helpful in revising the paper. Technical assistance was provided by R.E. Jones. This work was supported in part by NASA grant NAGW 3535.

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[*Manuscript received 27 January 1993:*
revised 14 September 1993]