The Fe-Ni metal equilibrium temperature of the unequilibrated Chainpur LL3.4 chondrite

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ABSTRACT: We report electron microprobe analyses of Fe, Ni and Co abundances in the associated α and γ phases in the Fe-Ni metal grains in unequilibrated ordinary chondrite Chainpur (type LL3.4). Our elemental measurements of five grains in chondrule and two grains in the matrix show that Co preferentially partitions into Kamacite relative to teanite, however the Co/Ni ratio in both phases is non-solar. The correlation of Co/Ni concentrations between associated kamacite and taenite in different metal grains emphasizes a common thermal event that established chemical equilibrium among individual metal grains and suggests that this chemical equilibrium of the metal grains could have been reached subsequent to chondrule formation events. Using the experimental study of Widge and Goldstein, the equilibrium temperature of these grains is 486±28°C; such a temperature is consistent with a post-accretion thermal metamorphism.

KEYWORDS: Fe-Ni, ordinary chondrite; temperature; metamorphism; chemical equilibrium.

RESUME: Nous présentons les analyses microsonde de l'abondance du Fe, Ni et Co dans les deux phases associées α (kamacite) et γ (taenite) des grains de métal FeNi dans la chondrite ordinaire inéquilibrée Chainpur (type LL3.4). Nos mesures montrent que le partage de Co se fait en faveur de la kamacite (phase α), mais le rapport Co/Ni dans les deux phases est non solaire ((Co/Ni)_{solaire}=0.045). La corrélation positive entre [Co]/[Ni] dans la kamacite et dans la taenite adjacente, dans les différents grains métalliques souligne un événement thermique commun qui a établi l'équilibre chimique entre les grains métalliques individuels et qui suggérer que cet équilibre chimique des grains du métal pourrait avoir été atteindre suite à l'événement de formation des chondres. Au moyen de l'étude expérimentale de Widge et Goldstein, la température d'équilibre de ces grains a été estimée à 486±28°C, température compatible avec un métamorphisme thermique post-accrétion.

MOTS-CLES : Fe-Ni, chondrite ordinaire, température, métamorphisme, équilibre chimique.

1. Introduction

The Fe-Ni metal is one of the major constituents in most chondritic meteorites [1]. It is generally believed that the Fe,Ni-metal formed in the solar nebula by high-temperature processes that included evaporation and condensation and and their iron and nickel alloys are a highly sensitive recorder of metamorphism in ordinary and carbonaceous chondrites [2, 3]. Studies of individual metal grains in several primitive chondrites classes [1, 4, 5, and 6] revealed that processes such as chondrule formation and parent body metamorphism were determinant in setting the final chemical compositions of its phases, kamacite and taenite.

In equilibrated chondrites the chemical composition of minerals is homogeneous in all parts of the meteorite; whereas in unequilibrated chondrites the metal grains exhibit varied chemical compositions. The partitioning of cobalt between kamacite and taenite can be used to determine the equilibration temperature of these phases. If these grains are brought to the same temperature, and if the equilibrium is achieved within single grains, equilibrium compositions of the kamacite and taenite must be expected. Based on experimental results [7], previous study of Co in the metal of

Bishunpur (LL3.15) revealed a correlation between adjacent kamacite/taenite phases [8] indicating metamorphic equilibration of this chondrite.

Here we present minor element (Co) measurements of associated kamacite and taenite in Chainpur (LL3.4 type), unequilibrated ordinary chondrites. Our aim is to shed light on chemical equilibrium temperature of this chondrite.

2. Exprimental procedures

2.1. Optical microscopy and metallography

A polished thin section of 86 mm² of the Chainpur (type LL3.4) unequilibrated ordinary chondrite was examined using optical microscopy and a JEOL JSM-840A Scanning electron microscope (SEM), at the Institut de Minéralogie, Physique des Matériaux et de cosmochimie in Paris, to locate suitable Fe-Ni metal grains with adjacent kamacite and taenite (α and γ phases) phases. The quantifications of major elements Fe and Ni in both α and γ phases in each individual Fe-Ni grain were performed using the Energy-Dispersive X–ray Spectrometry (EDX), equipped with a detector SD³ Bruker, at an accelerating voltage of 15kV and a working distance set at 15.7mm.

2.2. Electron microprobe analysis (EPMA)

Quantitative analyses of major and minor elements in metal (Fe, Ni and Co) were performed using the CAMECA SX-100 electron microprobe (EPMA) equipped with wavelength dispersive spectrometers at UPMC in Paris.

Measurements were performed at 15 kV accelerating voltage, using a beam current of 10 nA for Fe and Ni on one hand and 300 nA for Co on the other. The duration of counting for Fe, Ni and Co is 30s. These elements were analyzed sequentially on a LiF analyzer crystal.

To calibrate our measurements of Fe, Ni and Co we are used the following standards: Hematite (Fe_2O_3) , Nickel oxide (NiO) and Cobalt (Co) respectively.

Synthetic Fe-Ni alloys (0, 9.6, 17 and 30wt% Ni) were analyzed in order to improve the background correction to Co and to correct for the interference with Fe [9]. The contribution of $Fe_{K\beta}$ on the peak of Co is corrected by using the equation (1):

$$[Co]_{corr} = [Co]_{mes} - (0.0011 * ([Fe]_{mes} - 70) - 0.0835)$$
(1)

'corr' means the corrected concentration and 'mes' means the measured concentration.

Parameters were computed from a least square fit to the synthetic samples with a correction coefficient of 0.95. This correction is similar to that used previously [9] but the equation is slightly different according to instrumental differences.

Under these conditions, the detection limits are 0.1wt% for Fe, 0.14 wt% for Ni and 50 ppm (parts per million) for Co. Point analyses that had totals outside the range 97-102% were rejected. In addition a polished section of Coahuila iron meteorite was analyzed repeatedly during the analytical sessions in order of check the consistency of the correction method.

3. Results

By using the SEM, five chondrule metal grains (4cha, 4chb, 4chc, 5cha and 9cha) and two matrix metal grains (2grf and 3gra) are located in our section (see Fig.1). The cross-sectional areas of these grains varied between 1.5 and 10.6 μ m² and they are composed exclusively of associated kamacite and taenite (α and γ) without any intermediate oxidation boundary or other phase separating them, which ensures that predictable elemental diffusional exchange between the two phases was not subjected to any interruption except from temperature drop.



Figure 1: SEM BSE image of Chainpur section showing some of the investigated chondrules and the matrix metal grains. T: taenite, K: kamacite and Tr: Troilite (FeS).

In order to identify the compositions of the two phases composing the metal grains, we measured first the concentrations of the major elements Fe and Ni in these phases by Energy-Dispersive X–ray Spectrometry (EDX). An example of EDS spectra obtained by the analysis of the grains 4cha, 4chb and 4chc are shown in Figure 2. There we remark that the peak of the Ni is very high in taenite relative to the peak of Ni in the kamacite, which confirms the nature of these two phases.

These EDS measurements of the chondrules and matrix grains show that the Ni concentrations of the Ni in the kamacite of the chondrules grains and matrix grains vary between 3.5 and 5wt% in kamacite and 39.3 and 48wt% in taenite, which is consistent with the previous results of [10,11]. The Fe-Ni phases appear homogeneous since no chemical zoning is visible.

Using the electron micro probe analyzer (EMPA) analysis we measured the abundance of the major elements Fe, Ni and the minor element Co in the two adjacent phases for each selected Fe-Ni grains we localized before. The concentrations values of these elements are summarized in the Table 1:



Figure 2: EDS spectra obtained from the analysis of the kamacite and the taenite phases of the the grains: 4cha, 4chb and 4chc.

Table 1: Averaged chemical composition of Fe, Ni and Co, measured by EMPA, in kamacite and
taenite of the selected metal grain. All contents are by weight% (1σ is the error of the mean, i.e.
standard deviation/square root of N). N is the number of analysis per phase.

Grain	Area (μm^2)	Phases	Ν	Fe(%)	(±)1σ	Ni(%)	(±) 1σ	Co(%)	(±) 1σ	Total
4cha	3.96	Κ	12	95.94	0.07	03.68	0.06	0.25	0.03	99.86
		Т	5	59.73	5.69	40.00	5.72	0.12	0.03	99.84
4chb	1.44	K	11	95.97	0.05	03.61	0.06	0.25	0.04	99.83
		Т	2	51.36	0.25	48.25	0.28	0.22	0.05	99.84
4chc	1.53	K	6	95.89	0.09	03.59	0.10	0.40	0.04	99.87
		Т	4	51.87	0.91	47.70	0.92	0.26	0.02	99.83
5cha	1.57	K	8	95.07	0.74	04.46	0.76	0.35	0.03	99.87
		Т	5	57.34	5.77	42.29	5.78	0.17	0.02	99.80
9cha	1.68	K	16	94.96	0.07	04.47	0.08	0.41	0.04	99.84
		Т	3	60.21	4.14	39.38	4.14	0.19	0.02	99.77
2grf	0.65	K	6	95.38	0.18	03.57	0.17	0.92	0.08	99.86

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		Т	5	58.91	4.12	40.38	4.22	0.56	0.12	99.85
3gra	10.6	K	7	93.88	0.22	05.05	0.16	0.94	0.10	99.87
		Т	12	59.22	1.26	40.36	1.25	0.19	0.01	99.76

4. Discussion

The results described above which are consistent with [9], in showing that the Co in kamacite and taenite in chainpur exhibits a large range of chemical variations, which is in conformity with the unequilibrated nature of the chondrites [11, 12]. In Figure 3 we plotted the abundances of Co vs. Ni in kamacite (α) and taenite (γ). We note that the Co contents in kamacite are about twice higher than in taenite. According to [13] this result suggests that the equilibration temperature is not high.



Figure 3: Plot of Co versus Ni in metal grains. The points in black points represent the contents obtained by Affiattalab and Wasson (1977) [14]. The dashed line indicates the Co/Ni solar ratio from [15].

The affinity of the Co to kamacite can reasonably be explained by the fact that the Co atoms fit better in its *BCC* lattice than in the taenite *FCC* lattice. The partitioning of minor siderophile elements (like Co) is presumably controlled by the interplay of the size and electron configuration of the Co atoms and the crystal structures of kamacite and taenite [16].

The Co/Ni ratio in chondule grains kamacite is close to, but slightly above the solar value. In contrast this ratio in matrix kamacite it is more higher by nearly five time from the solar value. In the taenite grains this ratio is significantly lower than the solar value.

From mass balance calculations between kamacite and taenite in each grain, we find the bulk Co/Ni ratio in these metal grains in the range of 0.027 to 0.062, which are around the solar ratio value. These chemical compositions of the grains suggest that the metal grains could have formed from distinct precursors of solar compositions.

In Figure 4 we plotted the ratio of the $(Co/Ni)_{\alpha}$ versus the $(Co/Ni)_{\gamma}$ for each metal grains.



Figure 4: Plot of kamacite-vs-taenite (Co/Ni) ratios of single metal grains of Chainpur.

We see that the Co/Ni ratio in kamacite correlates with the Co/Ni ratio in associated taenite grains (Fig.4). The only point outside the correlation corresponds to the biggest grain; it's one of the matrix grains (3gra). We explain that this grain can come from another chondrite during the impact of the parent corps.

The correlations in Fig. 4 suggest that chemical equilibrium was reached between associated kamacite-taenite in each grain in Chainpur. We expect that this happened during a common thermal event. The $(Co/Ni)_{K}/(Co/Ni)_{T}$ trend gives a slope of 19.37 ± 5.78 with a coefficient of determination R^{2} value of 0.982.

According to [7], and using the equation determined by [17] we calculated an equilibrium temperature between kamacite and taenite of $486\pm28^{\circ}$ C.

Note that equilibration temperatures are determined from coexisting mineral phases and typically reflect the lowest temperature during the cooling of the system at which minerals remained in diffusive equilibrium [18]. This temperature is in the range of that reported by F.J.M.Reitmeijir et al. for the ordinary chondrite type 3.4–3.6 [19].

This temperature is consistent with a post-accretion thermal metamorphism and corresponds to the closure temperature for metal equilibration.

5. Conclusion

We have performed electron microprobe analyses of Fe, Ni and Co in associated kamacite and taenite in the LL3.4 ordinary chondrite Chainpur. Our main observations are the following:

1/ The elemental concentrations of Fe, Ni, and Co show large elemental variations among the metal grains, which is consistent with the unequilibrated character of the chondrite.

2/ Estimates of the bulk Co/Ni ratio of the metal grains show they are about solar values, which suggest that the metal grains could have formed from distinct precursors of solar compositions.

3/ The associated kamacite and taenite of the grains show correlated Co/Ni concentrations suggesting chemical equilibrium between the metal phases. This equilibrium was most likely established during a common thermal event subsequent to chondrule formation events.

4/ The calculated equilibrium temperature of the metal grains is 486±28°C; fairly consistent with a post-accretion thermal metamorphism for this equilibrium.

References

[1] Campbell, A. J., Zanda, B., Perron, C., Meibom, A., & Petaev, M. I.; *Chondrites and the Protoplanetary Disk*, 341, 407–430 (2005).

[2] Meibom, A., Desch, S. J., Krot, A. N., Cuzzi, J. N., Petaev, M. I., Wilson, L., & Keil, K.; *Science*, 288(5467), 839-841(2000).

[3] Kimura, M., Grossman, J. N., & Weisberg, M. K.; Science, 43(7), 1161-1177 (2008).

[4] Jacquet E., Paulhiac-Pison M., Alard O., and Kearsley A., *Meteoritics & Planetary Science*, 48, 1981–1999 (2013).

[5] Connolly H. C., Huss G. R. and Wasserburg G. J., *Geochimica et Cosmochimica Acta*, 65, 4567–4588 (2001).

[6] Hewins R. H., Yu Y., Zanda B., and Bourot–Denise M., *Antarct. Meteorite Res.* 10, 275–298 (1997).

[7] Widge, S. and Goldstein J. I, Met. Trans. 8A, 309-315 (1977).

[8] Mostefaoui S., Metal and carbon in primitive ordinary chondrites: Implications for the physicochemistry of the early Solar System, *Ph.D. thesis*. MNHN, Paris, France (1996).

[9] Afiattalab F. and Wasson J. T, *Geochimica et Cosmochimica Acta*, 44, 431–446 (1980). [10] Brearley A. J. and Rubin H. j., *Planetary Materials*, <u>Mineralogical Society of America</u>, New Mexico, USA (1998).

[11] Rambaldi E. R. and Wasson J. T., Geochimica et Cosmochimica Acta, 48, 1885–1897 (1984).

[12] Rubin A. E., Geochimica et Cosmochimica Acta, 54, 1217–1232 (1990).

[13] Goldstein J. I., Yang J., and Scott E. R. D., Meteoritics & Planetary Science, 47, 5132 (2012).

[14] Affiattalab F. and Wasson J.T., Geochimica et Cosmochimica Acta, (1977)

[15] Anders E. and Grevesse N., Geochimica et Cosmochimica Acta, 53, 179-214 (1989). [16] Hsu

W., Huss G. R., and Wasserburg G. J., Geochimica et Cosmochimica Acta, 64, 1133-1147 (2000).

[17] Wasson J. T., and Hoppe P., Geochimica et Cosmochimica Acta, 84, 508-524 (2012).

[18] Huss, Gary R., Alan E. Rubin, and Jeffrey N. Grossman, *Meteorites and the Early Solar System II*, 943, 567–586 (2006).

[19] Rietmeijer F.J.M. and Mackinnon I.D.R., Nature, 315, 733–736 (1985).