

**RE–OS ISOTOPE SYSTEMATICS AND FRACTIONATION OF SIDEROPHILE ELEMENTS IN METAL PHASES FROM CB CHONDRITES.** N. Nakanishi<sup>1</sup>, T. Yokoyama<sup>1</sup>, T. Usui<sup>1</sup>, and H. Iwamori<sup>1,2</sup>.

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**Introduction:** Metal is one of the main components of chondritic meteorites and a significant reservoir of Fe along with silicates and sulfides. Metal plays a key role in physicochemical processes that fractionate Fe (i.e., siderophile elements) from Si (i.e., lithophile elements) in the early solar system, generating variable chemical reservoirs before the onset of planetesimal formation. Therefore, the abundances of metal phases and the distribution of Fe among multiple phases are variable across different chondrite classes, which provide useful information regarding the origin and chemical state of the parent bodies of individual meteorites.

Highly siderophile elements (HSEs: Re, Os, Ir, Ru, Pt and Pd) have a great affinity for Fe-metal relative to silicate, which are very refractory and exist as gas only at high temperature. Therefore, HSEs in metals in a variety of meteorites can provide an important clue for understanding of high temperature processes in the nebula, as well as planetary differentiation. Specifically, the <sup>187</sup>Re–<sup>187</sup>Os isotope system yields chronological information regarding the fractionation of HSEs.

Numerous studies have conducted comprehensive analyses of HSE abundances in chondritic metals utilizing laser ablation ICP-MS (LA-ICP-MS) and instrumental neutron activation analysis (INAA) [e.g., 1–3]. Major findings of these studies include the relationship between HSE abundances and the size of metal grains in ordinary chondrites, formation processes of metal grains associated with chondrules in CR chondrites, and the origin of metal grains in metal-rich chondrites (CB, CH). However, these studies scarcely include in-situ Os isotope data due to analytical difficulties. We have developed a technique for in-situ measurement of Os isotopes in metal grains using a micro-milling system coupled with N-TIMS, and found that individual CB metals have <sup>187</sup>Os/<sup>188</sup>Os ratios close to the bulk CI chondrite value with limited variation [4].

This study is a follow up of our previous investigation that places emphasis on spot analyses of HSEs and other siderophile elements in CB metals where <sup>187</sup>Os/<sup>188</sup>Os ratios have been obtained. We utilize fs-LA-ICP-MS for conducting precise HSEs analysis in metal samples. By integrating overall measurements, we discuss the origin of metal grains in different types of CB chondrites.

**Experimental:** We examined multiple metal grains in three CB chondrites, Bencubbin (CB<sub>a</sub>), Gujba (CB<sub>a</sub>), and Isheyevu (CB<sub>b</sub>). The details for Os isotope

analysis using a micro milling system and N-TIMS are described in [4]. The concentrations of P, S, Cr, Fe, Co, and Ni in analytical spots adjacent to the sampling pits for Os isotope analysis were determined by EPMA (JEOL-JXA-8530F). The sample surface was polished and coated by carbon. All the analyses were performed with an accelerating voltage of 15 kV and a beam current of 10 nA. The standards were pure metals, apatite (P), celestite (S), Cr<sub>2</sub>O<sub>3</sub> (Cr), hematite (Fe), Co<sub>2</sub>SiO<sub>4</sub> (Co), and bunsenite (Ni) of known compositions. The concentrations of HSEs in analytical spots adjacent to the sampling pits were analyzed with fs-LA-ICP-MS (IFRIT, Cyber Laser). We ablated metal samples with fluences of 28 J cm<sup>-2</sup>, repetition rate of 180 Hz, pulse lengths of 220 fs, and wave lengths of 260 nm. We used an iron meteorite Chinga (IVB-an) as a standard in which concentrations of HSEs were calibrated using isotope dilution method. We used <sup>61</sup>Ni as an internal standard and normalized using the Ni content measured by EPMA for each micro milling spot.

**Results and Discussion:** Fig. 1 shows the <sup>187</sup>Re–<sup>187</sup>Os isochron for metal samples from CB chondrites. The data points are generally plotted on the 4.567 Ga Re–Os reference line calculated using the initial <sup>187</sup>Os/<sup>188</sup>Os isotope ratio (0.09517 at 4.567 Ga) obtained from IIIAB iron meteorites [5] and  $\lambda = 1.666 \times 10^{-11} \text{ y}^{-1}$  for <sup>187</sup>Re [6]. Nearly homogeneous <sup>187</sup>Os/<sup>188</sup>Os ratios in CB metals indicate that fractionation of Re and Os was minuscule during metal formation at ~4.57Ga. Because Re and Os are ultrarefractory elements with similar 50% condensation temperatures (Re: 1821 K, Os: 1812 K), the limited Re/Os variation may suggest simultaneous condensation of Re and Os from nebular gas during metal formation. Alternatively, several lines of evidence support the impact origin for CB metal grains [3,7]. Krot et al. [8] reported the age of CB chondrules from Gujba and HaH 237 by the <sup>207</sup>Pb–<sup>206</sup>Pb method, and concluded that the chondrules formed ~5 Myr after CAI condensation via planetary collision. Assuming that metal grains and chondrules from CB chondrites formed contemporaneously, our <sup>187</sup>Re–<sup>187</sup>Os data suggest that the redistribution of Re and Os during metal formation associated with planetary collision was not significant as are the cases of solidification of liquid metal.

Although the variation of Re/Os ratios in CB metal grains is limited, a signature of small Re–Os fractionation is recognized when initial Re/Os ratios calculated

from the  $^{187}\text{Os}/^{188}\text{Os}$  ratios are plotted against Os/Ir (Fig. 2). A positive correlation in this diagram for  $\text{CB}_a$  metal grains suggests small but discernable Re–Os fractionation along with Os–Ir fractionation. The ultra-refractory HSEs (Re–Os–Ir) do not fractionate during redox and sulfidation processes on the parent body, but can fractionate significantly via solidification of liquid metal [9]. However, the melting curves of an Fe–Ni metal with different S abundances trend toward opposite directions from  $\text{CB}_a$  metal data. Rather, the observation suggests that  $\text{CB}_a$  metal grain condensed either into solid metal from gas metal or into solid metal through liquid metal phase without keeping equilibrium condensation. We speculate that the condensation of metal grains occurred continuously at an equilibrium condition in a cooling gas until the condensation temperature of Ir ( $\sim 1600$  K), resulting in a systematic variation of Re/Os and Os/Ir ratios in  $\text{CB}_a$  metal grains.

Unlike ultra-refractory HSEs, fractionation among Pd, Fe, and Ni in CB metals is evident, which exhibits a positive correlation in the Pd/Fe vs Ni/Fe diagram (Fig. 3) as was observed in [3]. The slope of the correlation for our CB metals in this diagram is steeper than the theoretical fractionation curves representing equilibrium condensation from nebular gas at pressures of  $10^{-2}$ – $10^{-6}$  bar, suggesting the condensation of metal grains in an extremely high gas pressure ( $10^7 \times$  solar nebula). Such high pressure conditions could be produced by impact of planetary collision [3]. Interestingly, our data for metals from  $\text{CB}_b$  Isheyevu follow the trend of  $\text{CB}_a$  metals. This is inconsistent with the observation in previous studies [3,10,11] that Pd/Fe and Ni/Fe ratios in zoned and unzoned metal grains from  $\text{CB}_b$  chondrites (HaH 237, QUE 94627, and QUE 94411) are plotted on the equilibrium condensation curves at nebular pressures (Fig. 3). The new measurements conducted in this study indicate that metal grains from some  $\text{CB}_b$  chondrites, at least for Isheyevu, share a common origin of planetary collision as for the case of metal grains in  $\text{CB}_a$  chondrites Bencubbin and Gujba.

**References:** [1] Rambaldi E. et al. (1977) EPSL 36, 347–358. [2] Jacquet E. et al. (2013) Meteorit. Planet. Sci. 48, 1981–1999. [3] Campbell A. et al. (2002) GCA 66, 647–660. [4] Nakanishi N. et al. (2013) LPSC, abstract #2407. [5] Cook D. et al. (2004) GCA 68, 1413–1431. [6] Smoliar M. et al. (1996) Science 271, 1099–1102. [7] Rubin A. et al. (2003) GCA 67, 3283–3298. [8] Krot A. et al. (2005) Nature, 436, 989. [9] Chabot N. and Jones H. (2003) MAPS 38, 1425–1436. [10] Campbell A. et al. (2001) GCA 65, 163–180. [11] Campbell A. et al. (2005) MAPS 40, 1131–1148.

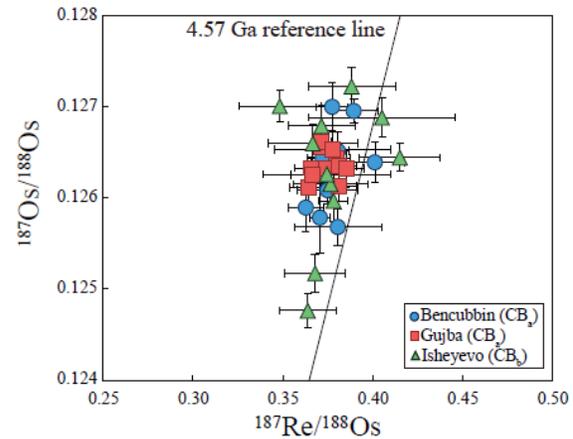


Fig. 1 Plots of  $^{187}\text{Re}/^{188}\text{Os}$  and  $^{187}\text{Os}/^{188}\text{Os}$  ratios for three CB chondrites analyzed in this study. The solid line is the 4.57 Ga reference line.

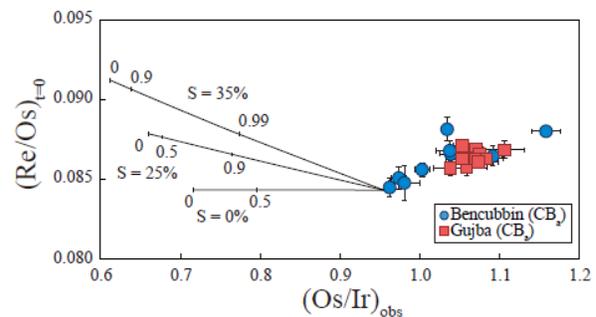


Fig. 2 Correlation between initial Re/Os ratios calculated from the  $^{187}\text{Os}/^{188}\text{Os}$  ratios and Os/Ir ratios in metal grains from Bencubbin and Gujba. Also shown are curves for batch melting of Fe–Ni metal, calculated using  $D$  values between solid metal and liquid metal.

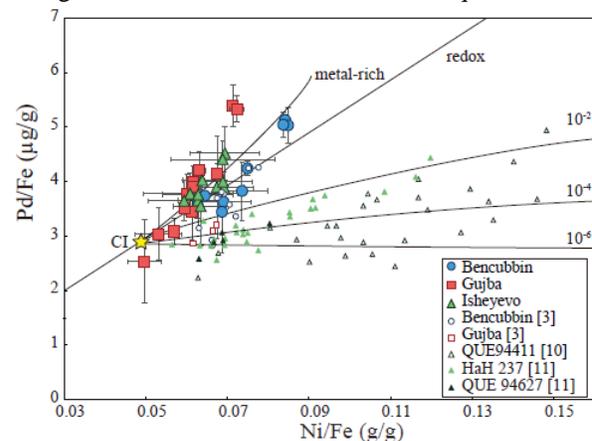


Fig. 3 Pd/Fe vs. Ni/Fe diagram for metal grains in CB chondrites. The right three curves are calculated equilibrium condensation trajectories at nebular pressures of  $10^{-2}$ ,  $10^{-4}$ , and  $10^{-6}$  bar. The metal-rich line is a condensation trajectory of metal enriched gas by a factor of  $10^7$  relative to the nebular conditions. The redox line represents the addition or removal of Fe during redox process [3].