

MOLYBDENUM ISOTOPIC EVIDENCE FOR NEBULAR THERMAL PROCESSING AND MATERIAL TRANSPORTATION IN THE INNER SOLAR SYSTEM. T. Yokoyama¹, Y. Nagai^{1,2}, R. Fukai¹ and T. Hirata²
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Introduction: The finding of nucleosynthetic isotope anomalies in meteorites overturned the traditional classification of meteorites. For instance, carbonaceous chondrites (CCs) and other meteorites (NCs: non-carbonaceous meteorites) are separately plotted in the $\epsilon^{50}\text{Ti}$ – $\epsilon^{54}\text{Cr}$ and $\Delta^{17}\text{O}$ – $\epsilon^{54}\text{Cr}$ spaces [1]. Importantly, the isotopic compositions of CCs and NCs are bimodal and form distinct clusters in these spaces. The CCs and NCs are also strongly bimodal in Mo isotopes. Recently, CCs and NCs are found to separately plot along two different lines (the CC- and NC-lines, respectively) in the $\mu^{95}\text{Mo}$ – $\mu^{94}\text{Mo}$ diagram [2–4], suggesting that the source materials feeding the CC- and NC-parent bodies were widely separated in the early Solar System.

To further investigate the Mo isotope dichotomy in meteorites, we obtained high-precision Mo isotope compositions in bulk aliquots of meteorites using negative thermal ionization mass spectrometry (N-TIMS). A specific emphasis was placed on chondritic meteorites of which the high precision Mo isotope data were scarcely obtained, while most of the recent high precision Mo isotope studies on meteorites focused dominantly on iron meteorites. With the extremely high precision Mo isotope data coupled with previously published data, we reevaluate the origin of Mo isotope anomalies in meteorites to better understand the formation history of materials accreted into meteorite parent bodies in the early Solar System.

Experiments: Meteorite samples used in this study are two CCs (CK4, CK5), five ordinary chondrites (OCs: H4, H5, LL5×2, LL6), two rumuruti chondrites (RCs: R3.9, R4), and ten irons (IIAB×4, IIE-like ungrouped, IVB×4, IVB-like ungrouped). Powdered samples of chondrites (~2 g) were dissolved with HF–HNO₃. All iron meteorites (~2 g) except the silicate-bearing irons were first leached in 6 M HCl and then digested in HNO₃–HCl. The silicate-bearing iron was first dissolved in HNO₃–HCl, then digested in HF–HNO₃. The subsequent chemical separation of Mo followed the method of [5]. Isotopic measurements of Mo were performed by an N-TIMS instrument (TRITON-plus, Thermo Fisher Scientific) installed at Tokyo Tech [6]. The mass fractionation was normalized by the exponential law that assumes $^{98}\text{Mo}/^{96}\text{Mo} = 1.453173$ in each sample. The Mo isotopic compositions are reported in $\mu^i\text{Mo}$ notation that is parts per 10⁶ deviations from the isotopic composition of the standard material (Kanto-Mo) measured in the same analytical campaign.

Results and Discussion: Figure 1 shows a $\mu^{95}\text{Mo}$ – $\mu^{94}\text{Mo}$ diagram of the chondrite and iron meteorite data obtained in this study. Also plotted are literature data for bulk meteorites published after 2015 [2–4,7]. The CK chondrites and IVB irons are plotted near the previously defined CC-line [4], while the other meteorites are consistently plotted on the previous NC-line [4]. This observation confirms the main conclusion of previous studies, namely, that the source materials feeding the CC- and NC-parent bodies were physically separated by a large distance in the early Solar System, presumably due to the formation of Jupiter’s core that opened a gap in the disk and prevented the exchange of materials between the two reservoirs [2]. In detail, however, some data points of the IVB irons form a linear trend of which the slope is distinct from those of the previous CC-lines. We consider that the original $\mu^i\text{Mo}$ values of some irons were modified by exposure to galactic cosmic rays. For this reason, we calculated the slopes and intercepts of the CC- and NC-lines by exclusively using Mo isotope compositions of bulk stony meteorites that were obtained in this study and in recent studies [2,3,7].

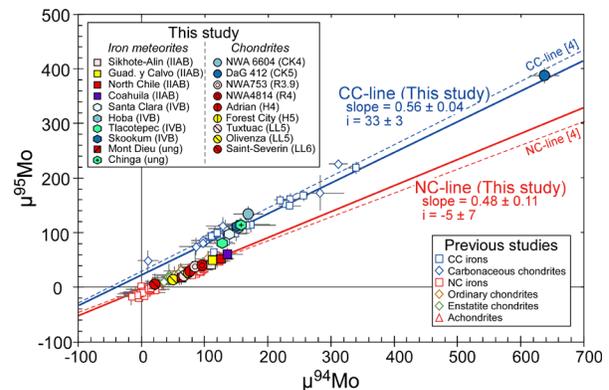


Fig. 1: $\mu^{95}\text{Mo}$ – $\mu^{94}\text{Mo}$ diagram of bulk meteorites obtained in this study and in previous studies [2–4,7]. Dashed lines are the previously defined CC- and NC-lines [4], and solid lines are those determined in this study by exclusively using bulk stony meteorite data.

In general, the CC-line in the $\mu^i\text{Mo}$ – $\mu^{94}\text{Mo}$ diagram has the same slope as the mixing line with an end-member component consisting of either mainstream presolar SiC grains [8] or the *s*-process component [9], suggesting that carriers enriched in *s*-process nuclides were involved in generating the Mo isotopic variabilities in CCs. In Fig. 1, the NC-line has a lower slope than that

of the CC-line, as well as that of the Earth vs. presolar SiC mixing line (not shown; slope = 0.54). Therefore, the NC-line represents a binary mixing between the Earth-like component and a component that cannot be explained by a pure s -process deficit relative to the Earth alone. To better understand the nucleosynthetic nature of the two end-components, we determined the enrichment coefficients of s - and r -process Mo isotopes (η_s and η_r) in each meteorite group (m) relative to a putative Solar System average material (SS_{ave}) represented by CI chondrite [11]. The η_s and η_r values are given by solving the following matrix equation;

$$\begin{pmatrix} \mu^{94}\text{Mo} \\ \mu^i\text{Mo} \end{pmatrix}_m = \begin{pmatrix} \mu^{94}\text{Mo} \\ \mu^i\text{Mo} \end{pmatrix}_{SS_{ave}} + \begin{pmatrix} -1 & 1 \\ -a_s^{i/94} & a_r^{i/94} \end{pmatrix} \begin{pmatrix} \eta_s \\ \eta_r \end{pmatrix}$$

where $i = 92, 95, 97, \text{ and } 100$, and $a_s^{i/94}$ and $a_r^{i/94}$ are the slopes of mixing lines between the coordinate ($\mu^{94}\text{Mo}$, $\mu^i\text{Mo}$) $_{SS_{ave}}$ and the pure s - and r -process end-member components, respectively. Note that $\mu^{94}\text{Mo}$ decreases and increases along with the addition of pure s - and r -process component, respectively.

In Fig. 2, the NC meteorites form a correlation trending from the Earth to the direction with lower η_s and η_r values, suggesting the presence of two end-member components NC-A and NC-B in the inner Solar System. Compared to NC-A, NC-B is depleted both in s - and r -process Mo isotopes. Most notably, the η_s and η_r values decrease in the order of Earth, enstatite chondrites (ECs), OCs, and RCs. In a recent evolutionary model of protoplanetary disk, Desch et al. [12] considered that the formation locations of chondritic parent bodies are 1.9–2.1, 2.2–2.6, and 2.6 AU for ECs, OCs, and RCs, respectively, which accounts for the abundances of CAIs and refractory lithophile elements in meteorites. Additionally, highly reduced and oxidized nature of ECs and RCs suggests that their parent bodies formed at a close and distant heliocentric distance in the inner Solar System, respectively. Consequently, the Mo isotopic composition in the NC region gradually changed from that of NC-A to NC-B as a function of heliocentric distance.

In the same evolutionary model [12], acapulcoites are thought to have formed in innermost part of the asteroid belt (~2.1 AU). However, as opposed to ECs, the acapulcoite-lodranite clan (Aca-Lod) is the most depleted in s - and r -process Mo isotopes among NCs (Fig. 2). We consider the inconsistency arises from the earlier accretion timing of the Aca-Lod parent body than the EC parent body (~1.3 and ~1.7 Myr after CAI, respectively; estimated by [12]). Therefore, the Mo isotopic composition in innermost Solar System gradually changed with time from that of NC-B to NC-A.

The above discussion leads us to conclude that the Mo isotopic composition in the inner Solar System was

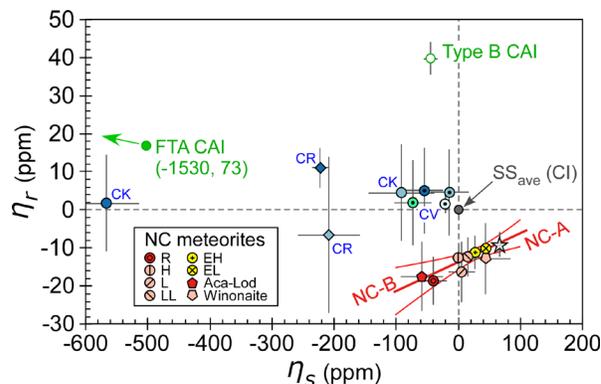


Fig. 2: Enrichment coefficients of Mo isotopes (η_s and η_r) in each meteorite group relative to the SS_{ave} (origin). Also shown are those of FTA and Type B CAIs [11, 13] and the Earth (star). Red line and curves are regression line and error envelopes for NCs + Earth, respectively

initially uniform as NC-B, followed by the incorporation of NC-A-like materials from the innermost Solar System by outward motion of particles due to turbulent diffusion in the disk. This scenario is consistent with that the Earth, which formed inner part and later timing than the parent bodies of NC chondrites, has the Mo isotopic composition most influenced by NC-A.

The origin of NC-A and NC-B reservoirs in the inner Solar System remains unclear. Assuming that the RCs are the closest representative of NC-B, this component is depleted only in r -process Mo relative to type B CAIs (Fig. 2). Therefore, NC-B could be formed by selective destruction of r -process-rich carrier via nebular thermal processing occurred in the disk with Mo isotopic composition slightly depleted in s -process than CI. On the other hand, the NC-A reservoir must have experienced a thermal processing with higher temperature at a closer heliocentric distance than the NC-B reservoir, resulting in the enrichments of s - and r -process Mo isotopes relative to NC-B. Interestingly, a fluffy type A (FTA) CAI [11] is significantly depleted in s -process Mo as opposed to NC-A, which may shed light on understanding the origin of NC-A reservoir in innermost Solar System.

References: [1] Warren, P.H. *EPSL* **311**, 93, 2011. [2] Budde, G. et al. *EPSL* **454**, 293, 2016. [3] Worsham, E.A. et al. *EPSL* **467**, 157, 2017. [4] Kruijjer, T.S. et al. *PNAS* **114**, 6712, 2017. [5] Nagai, Y. and Yokoyama, T. *Anal Chem* **86**, 4856, 2014. [6] Nagai, Y. and Yokoyama, T. *JAAS* **31**, 948, 2016. [7] Budde, G. et al. *GCA* **222**, 284, 2018. [8] Nicolussi G.K. et al. *GCA* **62**, 1093, 1998. [9] Bisterzo, S. et al. *ApJ*, **787**, 14pp, 2014. [10] Burkhardt, C. et al. *EPSL*, **357-358**, 298, 2012. [11] Burkhardt, C. et al. *EPSL*, **312**, 390, 2011. [12] Desch, S.J. et al. *ApJS*, **238**, 31pp, 2018. [13] Brennecka, G.A. et al. *PNAS*, **110**, 17241, 2013.