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#### **Key Points:**

- Planetary magmatic processes do not fractionate Ni stable isotopes
- Ureilite parent body (UPB) has similar δ<sup>60/58</sup>Ni values as chondrites, and Earth's core formation does not fractionate Ni stable isotopes
- Cr stable isotopes recorded partial melting and sulfur-rich core formation on UPB

#### **Supporting Information:**

Supporting Information may be found in the online version of this article.

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# Nickel and Chromium Stable Isotopic Composition of Ureilites: Implications for the Earth's Core Formation and Differentiation of the Ureilite Parent Body

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**Abstract** We report the first Ni and Cr stable isotope data for ureilite meteorites that are the mantle residue of a carbon-rich differentiated planet. Ureilites have similar Ni stable isotope compositions as chondrites, suggesting that the core-mantle differentiation of ureilite parent body (UPB) did not fractionate Ni isotopes. Since the size of Earth is potentially larger than that of UPB; with diameter >690 km), resulting in higher temperatures at the core-mantle boundary of Earth, it can be predicted that the terrestrial core formation may not directly cause Ni stable isotope fractionation. On the other hand, we also report high-precision Cr stable isotope composition of ureilites, including one ureilitic trachyandesite (ALM-A) that is enriched in lighter Cr stable isotopes relative to the main-group ureilites, which suggests that the partial melting occurred on UPB. The globally heavy Cr in the UPB compared to chondrites can be caused by sulfur-rich core formation processes.

**Plain Language Summary** The stable isotope fractionation of siderophile elements is robust to trace the planetary core formation processes. However, whether nickel (Ni) isotopes fractionate during the core formation is highly debated, since the origin of Ni stable isotope difference between bulk silicate Earth and chondrites is not clear. Here, we report high-precision Ni stable isotope data (expressed as  $\delta^{60/58}$ Ni, the per mil deviation of Ni<sup>60</sup>/Ni<sup>58</sup> ratios relative to NIST SRM 986) for ureilite meteorites that come from the mantle of a carbon-rich differentiated body. Ureilites have an average  $\delta^{60/58}$ Ni value of  $0.26 \pm 0.13\%$  (2SD, N = 22) that is highly consistent with that of chondrites with  $\delta^{60/58}$ Ni =  $0.23 \pm 0.14\%$  (2SD, N = 37), which suggests that planetary core formation does not effectively fractionate Ni stable isotopes. There is a ureilite trachyandesite that enriches lighter Cr stable isotopes ( $\delta^{53}$ Cr =  $-0.11 \pm 0.02\%$ c;  $\delta^{53}$ Cr as the per-mil deviation of Cr<sup>53</sup>/<sup>52</sup>Cr ratios relative to NIST SRM 979) relative to the main-group ureilites ( $\delta^{53}$ Cr =  $-0.05 \pm 0.04\%$ c; 2SD, N = 10), which suggests that the partial melting occurred on ureilite parent body (UPB). The globally heavy Cr in the UPB compared to chondrites can be caused by sulfur-rich core formation processes.

#### 1. Introduction

Core formation is one of the most significant stage of planetary formation and evolution (Jones & Drake, 1986), affecting the elemental composition of siderophile (metal-loving) elements. The effect of core formation on the composition of the silicate part of a planet can be reconstructed by using the stable isotope composition of siderophile elements (Bourdon et al., 2018). Nickel (Ni) is a major element in chondrites (>1 wt%), and ~90% of Ni budget is located into the core (Allègre et al., 1995; McDonough & Sun, 1995). In addition, Ni is a refractory (temperature of 50% condensation,  $T_{c50\%}$  of 1353 K (Lodders, 2003; P. A. Sossi et al., 2019), and mono-state ion (Ni<sup>2+</sup>) element in planetary mantles, so Ni stable isotopes mostly do not fractionate by volatile processes and magmatic evolution during planetary differentiation. Hence, Ni stable isotopes may be robust to trace planetary core formation by comparison between the compositions of planetary silicate fractions and chondrites that are taken as chemical proxies for bulk Earth and other terrestrial planets (Allègre et al., 1995), considering that the actual samples from the core are not accessible. Previous studies have comprehensively and systematically investigated the Ni stable isotope composition (expressed as  $\delta^{60/58}$ Ni, mass-dependent per mil deviation of Ni<sup>60</sup>/Ni<sup>58</sup> ratios relative to NIST SRM 986) of chondrites with an average  $\delta^{60/58}$ Ni value of 0.23  $\pm$  0.14% (2SD;

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Validation: Ke Zhu (朱柯), Frederic Moynier Writing – review & editing: Ke Zhu (朱柯), Jean-Alix Barrat, Akira Yamaguchi, Frederic Moynier Figure S1 in Supporting Information S1) (Cameron et al., 2009; Chernonozhkin et al., 2016; Gall et al., 2017; Klaver et al., 2020; Moynier et al., 2007; Wang et al., 2021) and of the bulk silicate Earth (BSE) with an average  $\delta^{60/58}$ Ni value of  $0.10 \pm 0.07\%$  (2SD) (Gall et al., 2017; Gueguen & Rouxel, 2021; Klaver et al., 2020; Saunders et al., 2020; Wang et al., 2021). Klaver et al. (2020) first observed the Ni isotope difference between chondrites and BSE, that is,  $\Delta^{60/58}$ Ni<sub>Chondrites-BSE</sub> =  $\sim 0.13\%$ , and interpreted it by terrestrial core formation. However, both *ab-initio* calculation (Guignard et al., 2020; Wang et al., 2021) and high-pressure experiments (Guignard et al., 2020; Lazar et al., 2012) show that core segregation may not effectively induce Ni stable isotope fractionation. Comparison of Ni stable isotope composition of chondrites and the silicate fractions of other planets in the Solar System is another way to test whether core formation fractionates Ni stable isotopes, and the key to unravel the Ni isotope paradox between chondrites and BSE. However, lack of Ni stable isotope data of other planets prohibits us testing this hypothesis.

Ureilite meteorites mostly represent mantle rocks of an early-differentiated carbon-rich planet (Berkley et al., 1980; Wilson et al., 2008; Zhu, Moynier, Schiller, Wielandt, et al., 2020), after the extraction of various types of magmas (Barrat et al., 2016; Bischoff et al., 2014; Cohen et al., 2004; Collinet & Grove, 2020) and a sulfur-rich iron melt (Barrat et al., 2015; Warren et al., 2006). Nickel is a compatible element that will reside in planetary mantles relative to crust; thus, the Ni isotope composition of main-group (unbrecciated) ureilites could be taken to represent that of bulk silicate ureilite parent body (UPB) (BSU = mantle + crustal reservoirs). As such, determining the potential Ni stable isotope difference between ureilites and chondrites could shed light on the core formation effect on Ni isotope fractionation, and further constrain the terrestrial core formation (Klaver et al., 2020). A similar approach using ureilites had been taken to study the effect of core formation on highly siderophile elements (Creech, Baker, Handler, et al., 2017; Creech, Moynier, & Bizzarro, 2017; Hopp & Kleine, 2021). Additionally, the size of UPB is smaller than Earth (Nabiei et al., 2018; Nestola et al., 2020; Schiller et al., 2018), and the effect of isotope fractionation during core formation for the UPB should be larger than that of the Earth due to higher temperature in the core-mantle boundary of larger bodies. More importantly, Ni is more siderophile with decreasing pressure (Bouhifd & Jephcoat, 2003), which means more Ni enters the core in small-size planets and Ni stable isotope composition of the silicate portion should be more remarkable for the smaller asteroids compared to Earth. Therefore, if no Ni isotope fractionation was detected during core formation on the UPB, it would represent an additional argument that terrestrial core formation would have not produced any detectable Ni isotope fractionation.

Chromium stable isotopes have also been widely used to study planetary differentiation (Bonnand, Parkinson,& Anand, 2016; Bonnand et al., 2020; Bonnand & Halliday, 2018; Jerram et al., 2021; Shen et al., 2020; P. Sossi et al., 2018; Zhu et al., 2019; Zhu, Moynier, Schiller, & Bizzarro, 2020; Zhu, Moynier, Alexander, et al., 2021; Zhu, Moynier, Schiller, Alexander, Barrat, et al., 2021); however, there is a lack of Cr isotope data for ureilites. Previous Cr isotopic observation about similar Cr stable isotope compositions between chondrites and BSE and data from high pressure experiments have proved that Cr isotopes do not fractionate during core formation (Bonnand, Williams, et al., 2016; Schoenberg et al., 2008; Zhu et al., 2019; Zhu, Moynier, Schiller, Alexander, Barrat, et al., 2021), except for sulfur-rich planets, that is, aubrites (Moynier et al., 2011; Zhu, Moynier, Schiller, Barrat, et al., 2021). Furthermore, the moderately volatile (Tc<sub>50%</sub> = 1291 K (Lodders, 2003; P. A. Sossi et al., 2019; Wood et al., 2019), and multi-valence-state (Cr<sup>2+</sup> and Cr<sup>3+</sup>) nature of Cr makes it a powerful tool to track planetary volatilization (P. Sossi et al., 2018; Zhu et al., 2019). In this way, investigating the Cr stable isotope compositions of UPB and comparing it with other planets/asteroids, including Earth (Schoenberg et al., 2008; P. Sossi et al., 2018), Moon (Bonnand, Parkinson, & Anand, 2016; P. Sossi et al., 2018), Vesta (Zhu et al., 2019), and enstatite achondrite parent bodies (Zhu, Moynier, Schiller, Barrat, et al., 2021), may provide insights into the differentiation history of UPB.

Here, we report high-precision Ni stable isotope data (using double spike techniques) for 22 ureilites and three chondrites to calibrate the core formation effect on Ni isotope fractionation and test whether the Ni stable isotope difference between chondrites and BSE could be due to core formation. We also report high-precision Cr stable isotope data (using double spike techniques) for 10 ureilites (mostly overlapping with the sample list for Ni stable isotope measurements) and one ureilitic trachyandesite (ALM-A) from Almahatta Sitta (Bischoff et al., 2014) to understand the origin and evolution of the UPB.

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Sample	Source	Fo (olivine core) (Fo%)	Ni content μg/g	$\delta^{60/58} \mathrm{Ni}~(\%e)$	2SD	N
Ureilites/Sahara						
NWA 2236	NIPR	96.8	1060	0.26	0.02	4
NWA 4471	JAB	78.1	3062	0.20	0.02	4
NWA 11372	UBO	77.8	77.8 926		0.05	4
NWA 11373	UBO	81.3	2360	0.29	0.05	4
NWA 5555	JAB	90.8	1381	0.28	0.03	4
NWA 5602	JAB	79	n.d.	0.31	0.02	4
NWA 5884	JAB	78.6 2272		0.21	0.02	4
NWA 6056	JAB	84.8 1269		0.31	0.05	4
NWA 7349	JAB	76.5 n.d.		0.17	0.05	4
NWA 7630	JAB	79.1	3381	0.24	0.06	4
NWA 7686	JAB	91	2248	0.37	0.06	4
NWA 7880	JAB	78.6	1005	0.24	0.03	4
NWA 8049	JAB	84.3	1403	0.24	0.07	4
Ureilites/Antarctica						
A 881931	NIPR	78.7	997	0.17	0.04	3
ALH 77257	NIPR	86.1	1236	0.22	0.03	4
ALH 82130	MWG	95.2	2110	0.25	0.02	4
EET 83225	MWG	88.3	459	0.36	0.03	4
LAP 03587	MWG	74.7	840	0.19	0.04	4
LAR 04315	MWG	81.9	1661	0.27	0.02	4
MET 01085	MWG	No olivine	429	0.41	0.05	4
Y 791538	NIPR	91.3	1472	0.36	0.03	4
Y 981810	NIPR	78.3	2074	0.26	0.02	4
Ureilite Average				0.26	0.13	2SD
Chondrites						
Allende (CV3)	USNM 3529		14760	0.23	0.02	4
Repeat				0.25	0.02	4
Orgueil (CI1):	MNHN 222		10800	0.14	0.05	4
Paris (CM2)	MNHN		14170	0.23	0.02	4
BIR-1	USGS		171	0.20	0.02	4

Note. NIPR: National Institute of Polar Research; JAB: Jean-Alix Barrat; UBO: Université de Bretagne Occidentale; MWG: NASA meteorite working group; USNM: National Museum of Natural History; Smithsonian Institution; Washington, DC; MNHN: Museum National d'Histoire Naturelle de Paris; USGS: United States Geological Survey.

## 2. Results

The detailed sample information, analytical methods, and data quality testing are described in Supporting Information S1, and the measured Ni and Cr stable isotope data are reported in Tables 1 and 2, respectively. There is no systematic  $\delta^{60/58}$ Ni difference between ureilites from Sahara Desert (0.26  $\pm$  0.11%, 2SD, N=13) and Antarctica (0.28  $\pm$  0.17%, 2SD, N=9). The  $\delta^{60/58}$ Ni variation for main group ureilites is small but clearly outside of the analytical uncertainty, ranging from 0.17  $\pm$  0.04% (A 881931 and NWA 7349) to 0.41  $\pm$  0.05% (MET 01085).

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Table 2
Cr Contents and Stable Isotope Compositions of Ureilites and Standards

Sample	Source	Mass mg	Fo (olivine core) (Fo%)	Cr content (Olivine core) µg/g	Cr content (Bulk) µg/g	<sup>55</sup> Mn/ <sup>52</sup> Cr	δ <sup>53</sup> Cr(‰)	2SD	N
NWA 2236	NIPR	n.d.	96.8	2600	n.d.	1.14	0.09	0.02	3
NWA 4471	JAB	9.9	78.1	3353	4575	0.73	-0.04	0.02	2
NWA 11372	ENS Lyon	9.3	77.8	3695	4502	0.78	-0.04	0.03	3
NWA 11373	ENS Lyon	7.7	81.3	4584	4051	0.68	-0.03	0.02	2
NWA 5555	JAB	8.5	90.8	4242	5109	0.78	-0.04	0.03	4
NWA 5602	JAB	11.7	79.0	4926	4984	0.70	-0.06	0.03	3
NWA 5884	JAB	9.8	78.6	4789	4662	0.74	-0.08	0.02	2
NWA 6056	JAB	13.0	84.8	5132	5381	0.72	-0.07	0.02	2
NWA 7630	JAB	15.0	79.1	5063	5355	0.62	-0.06	0.02	2
NWA 7686	JAB	12.6	91.0	4379	5231	0.76	-0.04	0.02	2
NWA 11368	JAB	7.8	96.8	n.d.	5261	0.77	-0.04	0.02	2
ALM-A	JAB	n.d.	no olivine	no olivine	1916	1.35	-0.11	0.02	2
NIST 3112a#	NIST						-0.04	0.02	25
BHVO-2#	USGS	~100					-0.11	0.02	1
PCC-1#	USGS	~100					-0.10	0.03	4

*Note.* n.d. not determined. The reference standard data marked with "#" measured in a same session have been reported in Zhu, Moynier, Alexander, et al. (2021). Data for Cr contents of olivine cores and Mn/Cr ratios are from Barrat et al. (2015) and Zhu, Moynier, Schiller, Wielandt, et al. (2020), respectively. Bulk Cr content of ALM-A is from Bischoff et al. (2014).

There are no clear relationships between  $\delta^{60/58}$ Ni values and Ni contents (Figure 1a;  $R^2 = 0.05$ ) or Mg#, that is, atom ratio of [Mg]/([Mg]+[Fe]), in the olivine core (Figure 1b;  $R^2 = 0.33$ ). It can be seen from the chemical maps (Figure S1 in Supporting Information S1) that Ni is mostly hosted together with Fe and S, and silicate minerals (e.g., pyroxene and olivine) are poor in Ni. All the main-group ureilites show an average  $\delta^{60/58}$ Ni of  $0.26 \pm 0.13\%$  (2SD),  $\pm 0.03\%$  (2SE, N = 22).

The  $\delta^{53}$ Cr values for the ureilites are also variable. In detail, NWA 2236 (with low Cr content in olivine core and high Mn/Cr ratio) has the highest  $\delta^{53}$ Cr value (0.09  $\pm$  0.02% $_{e}$ ), while ALM-A (with low Cr content and high Mn/Cr ratio) has the lightest  $\delta^{53}$ Cr value ( $-0.11 \pm 0.02\%e$ ), with the rest of main-group ureilites possessing homogeneous  $\delta^{53}$ Cr values of  $-0.05 \pm 0.04\%e$  (2SD, N=10). The  $\delta^{53}$ Cr values for the main-group ureilites do not correlate with their Cr contents, Fe/Mn ratios, and Mg# in the olivine cores (Figure 1). Also, the  $\delta^{53}$ Cr and  $\delta^{60/58}$ Ni values for the ureilites are independent of each other.

#### 3. Discussion

### 3.1. Nickel Stable Isotope Variation in Ureilites

Consistent  $\delta^{60/58}$ Ni values between ureilites from the Sahara Desert and Antarctica suggests that terrestrial weathering effect on Ni stable isotopes is limited. Lack of clear relationships between  $\delta^{60/58}$ Ni values and Ni contents and olivine core Mg# indicate limited Ni isotopic fractionation during igneous processes on the UPB. This is consistent with the fact that igneous processes do not effectively fractionate Ni stable isotopes on Earth, as evidenced by the absence of  $\delta^{60/58}$ Ni variations between peridotites  $(0.10 \pm 0.07\%)$  and basalts  $(0.03 \pm 0.16\%)$  (Klaver et al., 2020; Saunders et al., 2020, 2021; Wang et al., 2021). However, the variation of Mg# of ureilites may result from a mixing process, based on C,  $^{54}$ Cr, and noble gas isotope evidence (Barrat et al., 2017; Broadley et al., 2020; Zhu, Moynier, Schiller, Wielandt, et al., 2020), rather than smelting. This is consistent with the fact that silicate minerals in ureilites, that is, olivines and pyroxenes, do not dominate Ni (Figures S1 and S2 in Supporting Information S1).

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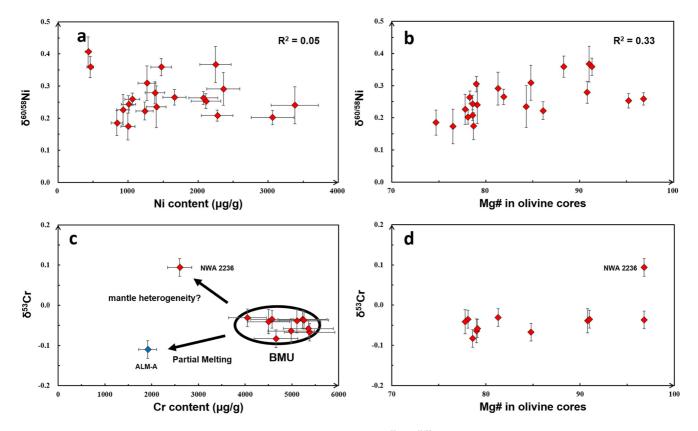


Figure 1. The relationships of Ni-Cr contents (bulk ureilites) and Mg# in olivine cores and  $\delta^{53}$ Cr- $\delta^{60/58}$ Ni values for ureilites (diamonds). The uncertainty for Ni and Cr contents are estimated as 10%. Figures 1a and 1b show that neither Ni contents nor Mg# in the olivine cores are not correlated to  $\delta^{60/58}$ Ni values ( $R^2 = 0.05$  and 0.33, respectively), suggesting magmatic processes do not fractionate Ni stable isotopes. In Figure 1c (BMU = bulk mantle of ureilite parent body(UPB), the Cr content for NWA 2236 is that of olivine core (Barrat et al., 2015) rather than bulk. The Cr elemental and isotopic difference between ALM-A (blue diamond) and the rest of maingroup ureilites (except for NWA 2236) can be caused by partial melting process, while the Cr depletion and enrichment of heavy Cr in NWA 2236 is possibly induced by mantle heterogeneity.

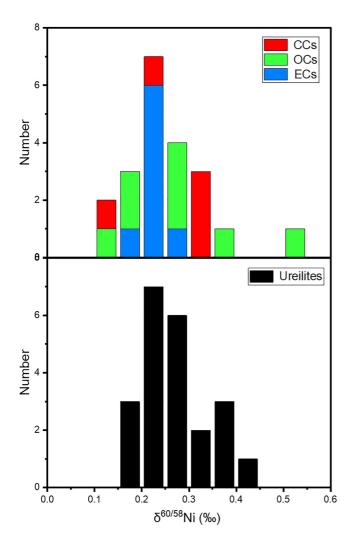
Although ureilites are mainly composed of olivine and pyroxene (ultramafic nature), Ni is mostly associated with Fe and S rather than Mg and budgeted in the minor accessory phases, for example, metal and sulfide with Ni contents of 1–6 wt% (Figures S1 and S2 in Supporting Information S1) compared to Ni contents of bulk ureilites of mostly 500–2,000 ppm (0.05–0.20 wt.%; Table 1). Since it has already been found that the sulfide can possess isotopically light Ni with  $\delta^{60/58}$ Ni down to  $\sim$ -1% (Gueguen & Rouxel, 2021; Hofmann et al., 2014), we interpret the small-scale  $\delta^{60/58}$ Ni heterogeneity in ureilites as reflecting various proportions of isotopically distinct sulfide (minor; mostly Ni<sup>2+</sup>) and metal (mostly Ni<sup>0</sup>). However, Ni contents in both metal and sulfide are variable (Figure S2 in Supporting Information S1), which might result in the lack of clear relationship between  $\delta^{60/58}$ Ni values and 1/[Ni] (Figure S3 in Supporting Information S1).

#### 3.2. Chondritic Nickel Stable Isotope Composition of Ureilite Parent Body Mantle

Since most of the Ni in ureilites is controlled by the metal phases that are accounting for up to 3%vol. in bulk ureilites (Goodrich et al., 2013) (Figures S1 and S2 in Supporting Information S1), the origin of the metal should be discussed. Most of the metal grains have largely fractionated highly siderophile elements relative to CV-CI chondrites, and require extremely high degrees (>98%) of batch Fe-S melt extraction (Goodrich et al., 2013). Additionally, during the breakup of the body and immediately after, carbon must have reacted with the olivines, producing their characteristic zonings (with Mg-rich rims), according to the reaction:  $C + MgFeSiO_4 = MgSiO_3 + CO + Fe$  (Warren & Huber, 2006). Therefore, most of the metals contained in the ureilites can be considered to be representative of the silicate mantle composition, instead of coming from the core.

The Ni stable isotope composition of UPB mantle can be estimated by the average  $\delta^{60/58}$ Ni values of all the main-group ureilites:  $0.26 \pm 0.13\%$  (2SD),  $\pm 0.03\%$  (2SE, N = 22), which is indistinguishable from that of

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**Figure 2.** Frequency distribution histogram of Ni stable isotopes for chondrites and ureilites. CC: carbonaceous chondrites, OC: ordinary chondrites and EC: enstatite chondrites. All the  $\delta^{60/58}$ Ni data for chondrites are listed in Table S1 in Supporting Information S1, and some of the data in the histogram overlap. Although there is a  $\delta^{60/58}$ Ni variation in both chondrites and ureilites, the variation range for them totally overlaps. This suggests that Ni stable isotopes do not fractionate during core formation of ureilite parent body (UPS).

chondrites  $\delta^{60/58}$ Ni = 0.23 ± 0.14% $_{o}$  (2SD) ± 0.02% $_{o}$  (2SE, N=37; Table S1 in Supporting Information S1) (Cameron et al., 2009; Chernonozhkin et al., 2016; Gall et al., 2017; Klaver et al., 2020; Moynier et al., 2007; Wang et al., 2021), including ordinary chondrites (OCs,  $\delta^{60/58}$ Ni = 0.25 ± 0.18% $_{o}$ , 2SD, N=16) that have close nucleosynthetic anomaly signatures (e.g., Ca<sup>48</sup>, Ti<sup>50</sup>, Cr<sup>54</sup> and Ni<sup>62</sup>) to ureilites (Quitté et al., 2010; Schiller et al., 2018; Trinquier et al., 2009; Yamakawa et al., 2010; Zhu, Moynier, Schiller, Wielandt, et al., 2020) (Figure 2). Thus, the UPB mantle has the same Ni stable isotope composition as UPB core, and core formation process for UPB did not fractionate Ni stable isotopes. This is in agreement with predictions from *ab-initio* calculation (Guignard et al., 2020; Wang et al., 2021) and high-pressure experiments (Guignard et al., 2020; Lazar et al., 2012).

Since core formation of the UPB, which must have occurred at a lower temperature (Barrat et al., 2015) than on Earth due to its likely smaller size, did not fractionate Ni stable isotopes, terrestrial core formation should also not fractionate Ni stable isotopes. Ni evaporation cannot be the cause of the isotopic fractionation neither, because (a) Ni is a refractory element (Lodders, 2003; P. A. Sossi et al., 2019); (b) Ni only has a single oxide, that is, NiO, compared to Cr that has multiple oxidized species (e.g., P. Sossi et al., 2018; Zhu et al., 2019), so both kinetic and equilibrium isotope fractionation during evaporation would enrich the residue (e.g., the BSE) in heavy Ni isotopes, which is opposite to what is observed as the BSE as a lower  $\delta^{60/58}$ Ni than chondrites.

Recently, Wang et al. (2021) proposed that the light Ni isotopic composition of the Earth was a consequence of the Moon forming a giant impact with a highly reduced planet with low- $\delta^{60/58}$ Ni value in the sulfur-rich mantle. Based on the siderophile element patterns (Warren et al., 2006), this hypothesis is also consistent with that UPB partitioned sulfur dominantly into the core, leaving a sulfur-poor mantle that has chondritic Ni isotope compositions. This hypothesis could be further tested, for example, via measuring the Ni stable isotope composition of aubrites and lunar samples. Alternatively, the inconsistent Ni isotopes between Earth and chondrites can simply indicate that Earth is not directly made by known bulk chondrites, which is also suggested by the recent radiogenic Cr isotopic (53Mn-to-53Cr decay system, with a half-life of 3.7 Ma) difference between Earth and chondrites (Zhu, Moynier, Schiller, Alexander, Davidson, et al., 2021). The Ni isotope compositions of chondrules deserve to be studied, since the pebble accretion model supports that chondrules can contribute to the accretional material of terrestrial planets (Johansen et al., 2015); especially the chondrules from enstatite chondrites that have similar isotope compositions of multiple elements as Earth's materials (Clayton et al., 1984; Steele et al., 2012; Trin-

quier et al., 2007; Zhu, Moynier, Schiller, & Bizzarro, 2020). Also Ni stable isotope fractionation can occur at the nebula stage, before accretion of UPB and other planets (Morbidelli et al., 2020).

# 3.3. Cr Stable Isotope Perspective on Differentiation of Ureilite Parent Body: Partial Melting, Core Formation, and Impact Processes

Except NWA 2236 ( $\delta^{53}$ Cr = 0.09  $\pm$  0.02% $_{e}$ ), all main-group ureilites show homogeneous  $\delta^{53}$ Cr values with an average of  $-0.05 \pm 0.04\%$  $_{e}$  (2SD, N=10) that are independent of Mg# in olivine cores (Figures 1c and 1d). This suggests a lack of Cr stable isotope fractionation during mantle processes in UPB. However, the  $\delta^{53}$ Cr values for ureilites are higher than that of the ureilitic trachyandesite, ALM-A, which has a  $\delta^{53}$ Cr value of  $-0.11 \pm 0.02\%$  $_{e}$  (Figure 1c). The only analyzed trachyandesitic composition (crustal sample) of the UPB, ALM-A, shows isotopically light Cr in relation to the main-group ureilites (mantle rocks), likely reflecting partial melting effect on Cr

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stable isotope fractionations (exchange of  $Cr^{2+}$  and  $Cr^{3+}$ ). This partial melting effect on Cr stable isotopes is also observed on Earth (Bonnand et al., 2020), Moon (P. Sossi et al., 2018), and Vesta (Zhu et al., 2019). Although C (Barrat et al., 2017),  $^{54}$ Cr (Zhu, Moynier, Schiller, Wielandt, et al., 2020), and noble gas (Broadley et al., 2020) isotope systems suggest that ureilite precursors were formed by mixing of two reservoirs, all the ureilites in this study (except NWA 2236) with variable Mg# and Fe/Mn ratios in the olivine cores have similar  $\delta^{53}$ Cr values, which indicates that the two reservoirs also have similar  $\delta^{53}$ Cr compositions. This is consistent with the fact that the different groups of chondrites have similar  $\delta^{53}$ Cr values (Zhu, Moynier, Schiller, Alexander, Barrat, et al., 2021).

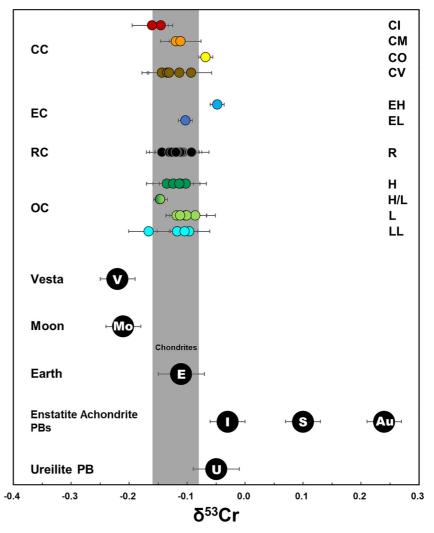
The size of UPB is highly debated, and the focus is on the origin of the microdiamond in the main-group ureilites (Nabiei et al., 2018; Nestola et al., 2020). Although some ureilites contain graphite and this graphite was transformed into diamond by shock (Nestola et al., 2020), some other diamonds cannot be produced upon impact (Nabiei et al., 2018). The fact that smelting failed to explain the huge composition range of olivines (Warren, 2012) and variation of C isotopes (Barrat et al., 2017) rules out definitively the possibility of a small UPB. Therefore, our following discussion will be based on a large-size UPB, for example, at least with diameter of ~690 km (Barrat et al., 2017; Warren, 2012), up to a Mars-sized planet (Nabiei et al., 2018).

Since ureilite are mantle rocks and also very rich in Cr (4,000–5,000 ppm; Table 2), the average  $\delta^{53}$ Cr value of  $-0.05 \pm 0.04\%$  (2SD, N = 10) for main-group ureilites (except NWA 2236) should represent that of bulk silicate UPB. Compared to chondrites that have homogeneous Cr stable isotope compositions, with  $\delta^{53}$ Cr =  $-0.12 \pm 0.04\%$  (2SD, N = 42) (Bonnand, Williams, et al., 2016; Schoenberg et al., 2016; Zhu, Moynier, Schiller, Alexander, Barrat, et al., 2021), the bulk silicate UPB possesses isotopically heavier Cr than chondrites that can represent precursor material of UPB (Figure 3), with  $\Delta^{53}$ Cr<sub>Ureilites-Chondrites</sub> =  $0.07 \pm 0.06\%$  (2SD) or  $\pm 0.02\%$  (2SE). This Cr stable isotope difference between bulk silicate UPB and chondrites can be attributed to the differentiation of UPB.

Chromium is a moderately volatile element ( $Tc_{50\%} = 1291$  K (Lodders, 2003; P. A. Sossi et al., 2019; Wood et al., 2019), and its isotopes fractionate during evaporation at planetary magma ocean stages, for example, Moon and Vesta (P. Sossi et al., 2018; Zhu et al., 2019). However, the heterogeneity in the  $\Delta^{17}$ O,  $\delta^{13}$ C, and  $\epsilon^{54}$ Cr values and noble gas isotope composition of these meteorites highly suggest that only a partial melting (as opposed to global scale magma oceans) occurred on UPB (Barrat et al., 2017; Broadley et al., 2020; Clayton & Mayeda, 1988; Zhu, Moynier, Schiller, Wielandt, et al., 2020). Therefore, the weak heating processes would have been unlikely to evaporate Cr that is not very volatile (as compared to e.g., Zn, Ga, Rb, and K). Additionally, the sulfur-rich core of UPB indicates a reduced condition during UPB differentiation, and the volatility of Cr decreases together with  $fO_2$  (P. A. Sossi et al., 2019), which is also consistent with the relatively high Cr abundance in ureilites (Table 2), in relation to chondrites with a Cr content of ~3,000 ppm (e.g., Alexander, 2019). This behavior of Cr makes it even less volatile than during the conditions prevailing for the magma ocean stages of Moon and Vesta (P. Sossi et al., 2018; Zhu et al., 2019) and limit any Cr loss by evaporation. Hence, we assume that the volatile process as the origin of the isotopically heavy Cr in bulk silicate UPB should be minor.

Chromium can be a siderophile during the core formation of large-sized planetary body, that is, at high temperature and pressure conditions (Siebert et al., 2013; Wood et al., 2008), which is consistent with the estimate that ~60% of terrestrial Cr entered the core (Allègre et al., 1995; McDonough & Sun, 1995). As for Earth's core formation, chondrites have similar Cr stable isotope compositions as BSE (Bonnand, Williams, et al., 2016; Schoenberg et al., 2008, 2016; Zhu, Moynier, Schiller, Alexander, Barrat, et al., 2021), suggesting no measurable Cr stable isotope fractionation occurred during terrestrial mantle-core differentiation, which is further supported by high-temperature and high-pressure experiments (Bonnand, Williams, et al., 2016). However, the partition coefficient of Cr into metal increases with the increasing sulfur (S) content (Bonnand & Halliday, 2018; Wood et al., 2014), but the experiments did not consider the compositional effect of S. Therefore, it does not necessarily apply to the scenario of sulfur-rich core formation process of the UPB (Warren et al., 2006). Note that the sulfurrich core formation may potentially cause the enrichment in the heavy Cr isotopes of aubrites (Zhu, Moynier, Schiller, Barrat, et al., 2021), and is also supported by ab-initio calculations (Moynier et al., 2011). Hence the higher  $\delta^{53}$ Cr value for bulk silicate UPB relative to chondrites can result from the S-rich core formation processes (Warren et al., 2006). Compared to the fractionation scale of aubrites, for example,  $\Delta^{53}$ Cr<sub>main-group aubrites-Chondrite</sub>  $_{s}=0.36\pm0.05~\%$ , the  $\Delta^{53}\mathrm{Cr}_{\mathrm{Ureilites-Chondrites}}=0.07\pm0.02~\%$  is much less, which may indicate a much larger size of UPB relative to that of main-group aubrite parent body, that is, <100 km (Wilson & Keil, 1991), since a

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# The δ<sup>53</sup>Cr Variation in Chondrites and Planets

Figure 3. Comparison of the  $\delta^{53}$ Cr variations among chondrites, achondrites, and the Earth-Moon system. The small colorful circles are chondrites, while the big black circles represent the Earth, Moon, Vesta, and enstatite achondrite and ureilite parent bodies. Abbreviation: CC-carbonaceous chondrites, OC-ordinary chondrites, EC-enstatite chondrites, V-Vesta, Mo-Moon, E-Earth, I-Itqiy, S-Shallowater, Au- main-group aubrites, and U-ureilites. The gray bar defines the average  $\delta^{53}$ Cr values ( $-0.12 \pm 0.04$ ; 2SD, N = 42) of all the chondrites. The  $\delta^{53}$ Cr difference between ureilites and chondrites results from a sulfur-rich core formation of ureilite parent body (UPB). Literature data sources: chondrites (Bonnand, Williams, et al., 2016; Schoenberg et al., 2016; Zhu, Moynier, Schiller, Alexander, Barrat, et al., 2021), Earth (Jerram et al., 2020; Schoenberg et al., 2008; P. Sossi et al., 2018), Moon (Bonnand, Parkinson, & Anand, 2016; P. Sossi et al., 2018), HEDs-Vesta (Zhu et al., 2019), and enstatite achondrites (Zhu, Moynier, Schiller, Barrat, et al., 2021).

higher pressure at planetary core-mantle boundary of a body would boost the isotope equilibrium between core and mantle and produce small isotope fractionation. The real sizes of the UPB and its core are unknown, so it is difficult to provide a quantitative model to test the hypothesis that Cr isotope fractionation occurred during core formation at present. The isotopically heavy  $Cr(\delta^{53}Cr = 0.09 \pm 0.02\%)$  in NWA 2236 is discussed in Supporting Information S1.

#### 4. Conclusions

1. Magmatic processes may not cause the fractionate Ni stable isotope variation in main-group ureilites, but the mixing of isotopically different sulfide and metal phases.

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- 2. Main-group ureilites have a similar  $\delta^{60/58}$ Ni value (0.26  $\pm$  0.13%; 2SD, N = 22) as chondrites (0.23  $\pm$  0.14%; 2SD, N = 37) that is consistent with the fact that core formation does not fractionate Ni stable isotopes. However, their Ni stable isotope difference possibly suggests that Earth did not originate directly from any of the known bulk chondrites.
- 3. Elevated Cr stable isotope compositions of main-group ureilites ( $\delta^{53}$ Cr =  $-0.05 \pm 0.04\%$ ; 2SD, N = 10) relative to the ureilitic trachyandesite (ALM-A;  $\delta^{53}$ Cr =  $-0.11 \pm 0.02\%$ ) and chondrites ( $\delta^{53}$ Cr =  $-0.12 \pm 0.04\%$ ) should result from partial melting and sulfur-rich core formation processes.

# **Data Availability Statement**

All of the original data have been deposited publicly to the repository of Zenodo (https://doi.org/10.5281/zenodo.5372803). All the supporting data can be found in the cited references (Barrat et al., 2015; Bischoff et al., 2014; Bonnand, Parkinson, & Anand, 2016; Bonnand, Williams, et al., 2016; Cameron et al., 2009; Chernonozhkin et al., 2016; Gall et al., 2017; Jerram et al., 2020; Klaver et al., 2020; Schoenberg et al., 2008, 2016; P. Sossi et al., 2018; Steele et al., 2012; Wang et al., 2021; Zhu et al., 2019, ; Zhu, Moynier, Schiller, Wielandt, et al., 2020; Zhu, Moynier, Schiller, Alexander, Barrat, et al., 2021; Zhu, Moynier, Schiller, Barrat, et al., 2021).

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