Tracing the origin and core formation of the enstatite achondrite parent bodies using Cr isotopes

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Abstract:

Enstatite achondrites (including aubrites) are the only differentiated meteorites that have similar isotope compositions to the Earth-Moon system for most of the elements. However, the origin and differentiation of enstatite achondrites and their parent bodies remain poorly understood. Here, we report high-precision mass-independent and mass-dependent Cr isotope data for 10 enstatite achondrites, including eight aubrites, Itqiy and one enstatite-rich clast in Almahatta Sitta, to further constrain the origin and evolution of their parent bodies. The ε^{54}Cr (per 10,000 deviation of the mass bias corrected ^{54}Cr/^{52}Cr ratio from a terrestrial standard) systematics define three groups: main-group aubrites with ε^{54}Cr = 0.06 ± 0.12 (2SD, N =7) that is similar to the enstatite chondrites and the Earth-Moon system, Shallowater aubrite with ε^{54}Cr = -0.12 ± 0.04 and Itqiy-type meteorites with ε^{54}Cr = -0.26 ± 0.03 (2SD, N =2). This shows that there were at least three enstatite achondrite parent bodies in the Solar System. This is confirmed by their distinguished mass-dependent Cr isotope compositions (δ^{53}Cr values): 0.24 ± 0.03 ‰, 0.10 ± 0.03 ‰ and -0.03± 0.03 ‰ for main-group, Shallowater and Itqiy parent bodies, respectively. Aubrites are isotopically heavier than chondrites (δ^{53}Cr =-0.12 ± 0.04 ‰), which likely results from the formation of an isotopically light sulfur-rich core. We also obtained the abundance of the radiogenic ^{53}Cr (produced by the radioactive decay of ^{53}Mn, $T_{1/2}$= 3.7 million years). The radiogenic ε^{53}Cr excesses correlate with the ^{55}Mn/^{52}Cr ratios for aubrites

(except Shallowater and Bustee) and also the Cr stable isotope compositions (δ^{53}Cr values). We show that these correlations represent mixing lines that also hold chronological significance since they are controlled by the crystallization of sulfides and silicates, which mostly reflect the main-group aubrite parent body differentiation at 4562.5 ± 1.1 Ma (i.e., 4.8 ± 1.1 Ma after Solar System formation). Furthermore, the intercept of these lines with the ordinate axis which represent the initial ε^{53}Cr value of main-group aubrites (0.50 ± 0.16, 2σ) is much higher than the average ε^{53}Cr value of enstatite chondrites (0.15 ± 0.10, 2SD), suggesting an early sulfur-rich core formation that effectively increased the Mn/Cr ratio of the silicate fraction of the main-group aubrite parent body.

1. Introduction

Enstatite achondrites, e.g., aubrites, are the only differentiated meteorite group that have similar oxygen isotope composition with that of the Earth-Moon system (Clayton et al., 1984; Greenwood et al., 2017). This isotopic similarity also extends to other isotope systems such as Ti, Cr or H, suggesting a close genetic link between the solid precursors of aubrites and the Earth-Moon system (Barrat et al., 2016; Greenwood et al., 2018; Greenwood et al., 2017; Newton et al., 2000; Piani et al., 2020; Trinquier et al., 2007; Zhang et al., 2012), although aubrites have distinct chemical and mineralogical composition and are more reduced than the Earth and the Moon (Keil, 2010a). Aubrites are more akin to the surface of Mercury (Ebel and Alexander, 2011; Nittler and Weider, 2019; Udry et al., 2019; Wasson, 1988). As such, the origin and evolution of aubrites may be closely linked to those of Earth, Moon and Mercury.

In detail, aubrites formed under highly reducing conditions with oxygen fugacities (fO_2) ranging from ~2 to ~6 log units below the iron-wustite (IW) buffer (Keil, 1968, 2010b). They consist predominately of FeO-poor enstatite (75 ~ 98% vol.) and contain minor silicate phases including plagioclase, diopside and forsterite (Watters and Prinz, 1979). Due to their formation under low fO_2 conditions, aubrites contain S-rich accessory minerals, such as troilite (FeS), alabandite (MnS) and oldhamite (CaS). Most aubrites are breccias and appear to originate from one or more parent bodies that underwent extensive melting and differentiation (Keil, 1989). Their highly siderophile

elements (HSE) depletion supports the idea that their parent body/bodies underwent core formation (van Acken et al., 2012a; van Acken et al., 2012b).

The formation of both aubrites and enstatite chondrites (ECs) occurred under extremely reduced conditions (Keil, 2010b; Lodders et al., 1993) and from precursors with similar isotopic compositions, suggesting a genetic relationship between the two parent bodies (e.g., Lodders et al., 1993; van Acken et al., 2012b). However, other lines of evidence including mineralogy, petrology, elemental abundances and cosmic ray exposure ages are difficult to reconcile with formation of aubrites from an enstatite chondrite precursor [see chapter 9.1 in Keil (2010b)]. Probing their relative genetic relationships may be improved by a better understanding of the chronology of formation of the aubrites, which is currently poorly defined. The lack of a radiogenic ^{26}Mg deficit in aubrites, while they have sub-chondritic ^{27}Al/^{24}Mg ratios, suggests that they formed after the decay of ^{26}Al, i.e. at least 3 million years (Ma) after Solar System formation (Baker et al., 2012). Iodine-Xenon systematics of Shallowater suggests it formed at 4562.4 ± 0.2 Ma (Pravdivtseva et al., 2017). Given that Shallowater is an unusual enstatite achondrite, which likely originates from a distinct parent body relative to other aubrites (Keil, 2010b; Moynier et al., 2011a), the timing of formation of Shallowater cannot be generalized to the main-group aubrites. Thus, the timing of aubrites and aubrite parent body formation remains largely unconstrained.

Chromium isotope systematics of aubrites may improve our understanding of the origin of aubrites. 1) The abundance of the neutron-rich isotope ^{54}Cr (nucleosynthetic)

varies among different meteorites (Trinquier et al., 2007). This feature, expressed as the ε^{54}Cr (per 10,000 deviation of the mass bias corrected ^{54}Cr/^{52}Cr ratio from to the terrestrial standard NIST SRM 979) signature, has been established as a tracer of the genetic relationships between meteorites (Qin et al., 2010; Trinquier et al., 2007; Zhu et al., 2021b). 2) The radiogenic isotope ^{53}Cr is the daughter product of the short-lived isotope ^{53}Mn (half-life of 3.7 million years (Holden, 1990). This decay system can provide precise relative chronology of early Solar System events while ^{53}Mn was still present (Birck and Allègre, 1988; Lugmair and Shukolyukov, 1998). The ^{53}Mn-^{53}Cr chronometer is especially sensitive to volatilization and magmatic processes, because Mn and Cr possess different volatility and chemical behavior during magmatic differentiation (Lodders, 2003; Sossi et al., 2019). This system has been successfully applied to date the differentiation of other planetary objects, including: Vesta (Lugmair and Shukolyukov, 1998; Trinquier et al., 2008b), angrite (Zhu et al., 2019b) and ureilite parent bodies (Van Kooten et al., 2017; Yamashita et al., 2010; Zhu et al., 2020b). 3) Mass-dependent Cr stable isotope variations (expressed as δ^{53}Cr, the per mil variation of the ^{53}Cr/^{52}Cr ratio relative to the terrestrial standard NIST SRM 979) can be used to trace planetary processes, such as volatile depletion (Sossi et al., 2018; Zhu et al., 2019c), magmatic differentiation (Bonnand et al., 2020a; Bonnand et al., 2020b; Bonnand et al., 2016a; Shen et al., 2019; Sossi et al., 2018; Zhu et al., 2019c), and possibly core formation (Moynier et al., 2011b).

So far, the only ε^{54}Cr value of aubrites has been reported for Bishopville (-0.16 ±

0.19) (Trinquier et al., 2007). This isotopic composition overlaps with the ε^{54}Cr signatures of enstatite (0.02 ± 0.11; 2SD, N = 13), Rumuruti (-0.06 ± 0.08; 2SD, N = 12) and ordinary (-0.39 ± 0.09; 2SD, N = 18) chondrites (Mougel et al., 2018; Pedersen et al., 2019; Qin et al., 2010; Trinquier et al., 2007; Zhu et al., 2021a; Zhu et al., 2021b). Consequently, the currently available ε^{54}Cr datum for aubrite limits its use to test genetic links to chondrites (e.g., enstatite and Rumuruti chondrites with ε^{54}Cr of ~0) and other achondrites (e.g., martian meteorites and angrites with ε^{54}Cr of ~-0.2 and ~-0.4 respectively; Trinquier et al., 2007; Zhu et al., 2019b), as well as possible mantle heterogeneity of the aubrite parent body and whether there are multiple parent bodies for enstatite achondrites (e.g., Zhu et al., 2020b). Shukolyukov and Lugmair (2004) reported ε^{53}Cr values for three bulk aubrites that established a ^{53}Mn-^{53}Cr isochron corresponding to a ^{53}Mn/^{55}Mn initial ratio of (2.0 ± 0.5) × 10^{-6} and an absolute age of 4560.8 ± 1.4 Ma, relative to the U isotope corrected U-Pb age of the D'Orbigny angrite (Amelin, 2008; Brennecka and Wadhwa, 2012; Glavin et al., 2004). Although the limitation of a three-point isochron makes the age significance of the isochron somewhat uncertain, these data provide promising evidence that Mn-Cr dating could fruitfully be used to better constrain the timing of aubrite formation and differentiation. Combined with Cr stable isotope information, these data may shed more light on the magmatic evolution and differentiation history of the aubrite parent bodies.

Aubrites are not the only class of enstatite achondrites. Recently, Harries and

Bischoff (2020) observed a new type of enstatite-rich differentiated meteorite clasts in Almahatta Sitta, which are believed to come from another asteroid with a size of ~500 km. These clasts also show petrological affinity to Itqiy, an EH7-anomalous meteorite, as they both underwent partial melting and are petrologically different from aubrites (Keil and Bischoff, 2008; Moynier et al., 2020; Patzer et al., 2001). The Cr isotope compositions of these different types of enstatite achondrites can be used to test their relationship to each other and to enstatite chondrites. Thus, here we report high-precision mass-independent and mass-dependent Cr isotope data for eight aubrite meteorites, the Itqiy meteorite and one enstatite achondrite clast from Almahatta Sitta.

2. Samples and analytical methods

2.1 Sample description.

The eight aubrites selected for this study are Khor Temiki, Bustee, Norton County, Allan Hills [ALH] A78113, ALH 84007, Larkman Nunatak [LAR] 04316, Shallowater and Cumberland Falls. We also studied two other enstatite-rich achondrite meteorites, Itqiy (EH7-anomalous) and one enstatite-rich clast in Almahatta Sitta, MS-MU-19 (Harries and Bischoff, 2020). Itqiy and MS-MU-19 are enstatite-rich rocks, both of which lack plagioclase and high-Ca pyroxene, suggesting they underwent higher degrees of partial melting. Their detailed petrology are described in Patzer et al. (2001) and Harries and Bischoff (2020), respectively. The fractions of Itqiy and MS-MU-19 that we used had been previously partially processed and leached by hot (120°C) 6N HCl in order to remove metal and sulfides for trace element studies. While a fraction of the Cr must have been lost during this process, the Cr of the acid residues that we obtained (mostly from the silicates) likely represent the primitive ε^{54}Cr value, since enstatite is the major Cr-host of these samples and magmatic process would have had homogenized the ε^{54}Cr signatures at the meteorite scale in any cases. We also dissolved a bulk Itqiy from another chip for mass-dependent Cr isotope study.

2.2 Sample dissolution

Small fragments of eight aubrite meteorites were crushed to powders using an agate mortar. From this powdered material, 30–50 mg were transferred into Teflon

bombs for digestion. We also dissolved a small chip (26.4 mg) of LAR 04316 for a replicate. Samples were dissolved following the protocol described in Inglis et al. (2018) using Teflon bombs and an Analab EvapoClean, which has been successfully applied in previous Cr isotopic studies of meteorites (Zhu et al., 2019b; Zhu et al., 2020b). The procedure involved heating in concentrated HF and HNO_3 (2:1) at 140 °C for two days, and subsequent dissolution in 6N HCl (also at 140 °C) for another two days to ensure complete digestion of fluorides, and refractory phases such as chromite and spinel. After complete digestion a ~2% aliquot was used for determination of the $^{55}Mn/^{52}Cr$ ratios and major element contents by ICP-MS in IPGP, ~50% of the digestion were purified for Cr by the following four-step column chemistry described in (Zhu et al., 2021b) for mass-independent Cr isotopic analyses, and the residual 40% were spiked with a ^{50}Cr-^{54}Cr double spike before a two-step column Cr purification for mass-dependent Cr isotopic analyzes of which method is described in the literature (Sossi et al., 2018; Zhu et al., 2021a; Zhu et al., 2019c).

2.3 Mass–independent Cr isotope ($\varepsilon^{53}Cr$ and $\varepsilon^{54}Cr$) analyses

We first used an anion chromatographic purification scheme to efficiently remove Fe from the remaining sample in 6M HCl, followed by elution of Cr on a 1-mL cation exchange column in 20 mL of 0.5M HNO_3 to remove the major elements including Mg, Ca, Al, Ni (Bizzarro et al., 2011) and collect all the Cr species (major Cr^0 and minor Cr^{2+} and Cr^{3+}) to reach a ~100% recovery. Prior to sample loading on the cation column, we used a Cr pre-treatment procedure involving dissolution in 10M HCl at >120 °C to

186 efficiently promote the formation of Cr(III)-Cl species, which have a low affinity for
187 the cation exchanger and thus elutes early (Larsen et al., 2016; Trinquier et al., 2008a).
188 The third clean-up column involved Cr purification from Al, Fe, V, Ti (and other
189 high-field-strength elements) and Na, K on a small (0.33ml) cation exchange column
190 using 0.5M HNO_3, 1M HF and 6M HCl (Larsen et al., 2018). Prior to sample loading
191 onto this last column, we used a Cr pre-treatment procedure involving exposure to
192 0.5M HNO_3 + 0.6% H_2O_2 at room temperature for >1 day to promote the formation of
193 Cr^{3+} (Larsen et al. 2016). However, it is difficult to transform all the Cr to Cr^{3+}, so we
194 collect the Cr^0 in 0.5 ml solution of sample loading and 0.5 mL 0.5N HNO_3 elution to
195 increase the recovery to >95% in this column. Finally, in the fourth column, 0.7 mL of
196 TODGA resin were used in 8N HCl to remove the residual Fe, V and Ti which are
197 isobars for ^{54}Cr (^{54}Fe) and ^{50}Cr (^{50}V and ^{50}Ti) (Pedersen et al., 2019). The four columns
198 can reach a total yield between 95% and 99%, and effectively remove Fe, V and Ti,
199 which benefits the isotope analysis on MC-ICPMS. The total blank of <5 ng is
200 negligible compared to the 5-20 μg of Cr processed through the columns. The final Cr
201 solution was re-dissolved in 100μL of concentrated HNO_3 and then dried again 2-3
202 times to transform the acid media and to minimize residual organics (i.e. from the
203 cation exchange resin).

204 The $^{55}Mn/^{52}Cr$ ratios with an uncertainty of 5% were measured on a Neptune plus
205 multiple-collector inductively-coupled-plasma mass-spectrometer (MC-ICP-MS)
206 following the protocol described by (Zhu et al., 2021b) and previously used in (Zhu et

al., 2019b; Zhu et al., 2020b). The Cr isotopic compositions were determined using a Neptune Plus MC-ICP-MS located at the Centre for Star and Planet Formation, Globe Institute, University of Copenhagen. Detailed analytical and data reduction method are described in (Zhu et al., 2021b). Sample solutions with concentration of less than 1 ppm were introduced via an ESI APEX and analyzed at a ^{52}Cr signal of 30-40 V, except for Norton County, with low-Cr content (180 ppm), that was analyzed at 15 V. Each sample was measured five times by sample-standard bracketing relative to the NIST SRM 979 Cr standard. The ^{53}Cr/^{52}Cr and ^{54}Cr/^{52}Cr ratios were normalized to a constant ^{50}Cr/^{52}Cr ratio of 0.051859 using an exponential law (Lugmair and Shukolyukov, 1998) and are expressed in the epsilon notation:

$$\varepsilon\,{}^{x}Cr = \left(\frac{({}^{x}Cr/{}^{52}Cr)_{sample}}{({}^{x}Cr/{}^{52}Cr)_{NIST\ SRM\ 979}} - 1\right) \times 10000 \quad (1),$$

with x = 53 or 54.

The measurements in this study are performed following Zhu et al. (2021b), and the related elemental doping tests (for Ti, V, Fe, K, and Na), external precision tests and the isotope data of known standard samples for data quality (Allende, DTS-1, PCC-1, DTS-2b) can be also found in Zhu et al. (2021b). The data for the reference standards are consistent with previously reported values (Mougel et al., 2018; Qin et al., 2010; Schiller et al., 2014; Schneider et al., 2020; Trinquier et al., 2007; Trinquier et al., 2008b; Zhu et al., 2019a; Zhu et al., 2019b; Zhu et al., 2020b), confirming the accuracy of our new data.

For acid leachates of Itqiy and the enstatite achondrite clast from Almahatta Sitta (MS-MU-19), the purification followed a two-step cation column as originally described in Trinquier et al. (2008a) and modified in Zhu et al. (2019c). Low-yield (e.g., < 70%) Cr purification may produce large equilibrium mass-dependent Cr isotope fractionation from column chemistry which cannot be well corrected using the exponential law (Qin et al., 2010; Trinquier et al., 2008a). Thus, similar to the method used in Moynier et al. (2011b), we used 3 (1 + 1 + 1) mL of 6M HCl for washing the column and we collected the residual material from the first 1-mL cation column and re-passed them in the first column to ensure yields ranging from 88% to 99% (average of 93%). The blank of the full chemical purification is between 0.5 and 2 ng and is negligible compared to 2 – 4 μg of Cr processed through the purification scheme. The mass-independent Cr isotope analysis was obtained via the total evaporation method on the Triton TIMS housed at Freie Universität Berlin based on methods described in the literature (Van Kooten et al., 2016; Zhu et al., 2019b; Zhu et al., 2020a). NIST 3112a was used as the isotope standard, because there is no resolvable mass-independent Cr isotope difference between NIST SRM 979 and NIST SRM 3112a (Zhu et al., 2021b). Using this method, we also measured the Cr isotope composition from the same dissolution of the Allende meteorite and DTS-1 (terrestrial dunite, USGS terrestrial standard) samples that were used for the MC-ICP-MS analysis, and the data are consistent. The data for individual measurements of TIMS total-evaporation measurements are listed in the Appendix.

2.4 Mass–dependent Cr isotope (δ^{53}Cr) measurements

The procedure for mass–dependent Cr isotope analysis was performed at the IPGP on aliquots of the same dissolutions used for mass–independent isotope measurements. First, the appropriate amount of ^{50}Cr–^{54}Cr double spike (28% of the Cr content endemic to the sample) was added to the aliquot, which was then refluxed in a closed beaker at 120 °C overnight to homogenize the sample and spike (e.g., Wu et al., 2020). The Cr of the samples was chemically purified via a typical two–step cation exchange chromatography (Sossi et al., 2018; Zhu et al., 2021a; Zhu et al., 2019c), modified from Trinquier et al. (2007). This method has a total procedural yield of 60–90% and a blank of ~5 ng of Cr. Compared to the total 3-40 µg of Cr from the samples, the blank can be neglected. The final Cr cut was evaporated in concentrated HNO_3 drops 3 to 5 times to convert the HCl medium to HNO_3 and to remove leftover organics (i.e., the resin) following which the samples were diluted to a concentration of 1 ppm Cr, in 2% (0.317 M) HNO_3 for isotope analysis.

The Cr stable isotope compositions of these purified samples were measured on a Thermo Scientific Neptune Plus MC–ICP–MS housed at the IPGP. Analytical details are described in Zhu et al. (2021a), Sossi et al. (2018) and Zhu et al. (2019c). The purified sample solution was diluted to ~ 1ppm, which returned a signal of ~ 15V of ^{52}Cr with a Stable Introduction System (SIS). Every measurement is composed of 100 cycles, with integration time of 4.194 s for each cycle. The final Cr stable isotope data

(δ^{53}Cr) for aubrites were corrected using their respective mass–independent Cr isotope compositions that had previously been analyzed on the same aliquots. The isotopic ratio for samples is reported in delta notation relative to NIST SRM 979:

$$\delta^{53}Cr(‰) = \left(\frac{(^{53}Cr/^{52}Cr)_{sample}}{(^{53}Cr/^{52}Cr)_{NIST\ SRM\ 979}} - 1\right) \times 1000 \quad (2)$$

The uncertainties quoted are the 2SD of individual sample measurements or the 2SD reproducibility of several NIST SRM 979 measurements in the same analytical session (~0.03 ‰), whichever is the largest. Similarly, the Cr stable isotope compositions of standard samples, including NIST SRM 3112a (unpurified, δ^{53}Cr = -0.08 ± 0.03 ‰; 2SD, N = 5), DTS-1 (δ^{53}Cr = -0.03 ± 0.03 ‰; 2SD, N = 5), PCC-1 (δ^{53}Cr = -0.04 ± 0.03 ‰; 2SD, N = 6) and BHVO-2 (δ^{53}Cr = -0.15 ± 0.03 ‰; 2SD, N = 2), were also measured in this study to test the data quality. Our data are consistent with previous studies (Bonnand et al., 2016b; Liu et al., 2019; Schoenberg et al., 2016; Sossi et al., 2018; Wu et al., 2020; Zhu et al., 2019c), which suggests that our δ^{53}Cr data are accurate to the stated uncertainties.

3. Results

All the sample information, elemental contents, mass-independent (ε^{53}Cr and ε^{54}Cr) and mass-dependent (δ^{53}Cr) Cr isotope data together with literature Cr and O isotope data from Shukolyukov and Lugmair (2004), Barrat et al. (2016), Clayton et al. (1984) and Newton et al. (2000) are reported in Table 1. The ε^{54}Cr variability between the analyzed aubrites is rather limited with six out of the nine aubrites (including two LAP 04316) having a mean ε^{54}Cr = 0.04 ± 0.07 (2SD, N = 6; Figure 1). The two aubrites with the extreme ε^{54}Cr values are Shallowater (-0.12 ± 0.04) and Khor Temiki (0.18 ± 0.04). Compared to aubrites, Itqiy and the enstatite achondrite clast in Almahatta Sitta, MS-MU-19, have lower ε^{54}Cr values, averaging at -0.26 ± 0.03 (2SD, N =2).

The mass-dependent Cr isotope data (δ^{53}Cr) for aubrites range from 0.10 ± 0.03 ‰ (Shallowater) to 0.30 ± 0.03 ‰ (Bustee), and bulk Itqiy possesses δ^{53}Cr of -0.03 ± 0.03 ‰. The relationship between the δ^{53}Cr and ε^{54}Cr values of these enstatite achondrites are shown in Figure 2a. Among main-group aubrites, δ^{53}Cr values broadly increase with bulk Mg# (Mg/(Mg+Fe)×100%, atom ratios; > 98; Figure 2b), but there is no correlation between their δ^{53}Cr values and bulk Cr contents (Figure 2c). The δ^{53}Cr values of main-group aubrites are, however, correlated with iron isotope compositions [expressed as δ^{56}Fe; (Wang et al., 2014)] (Figure 2d). Itqiy, Shallowater and main-group aubrites also have different and correlated Mg# and δ^{53}Cr values

(Figure 2b and Figure 3).

The measured ε^{53}Cr values range from 0.00 ± 0.02 for Shallowater up to 2.41 ± 0.03 for Bustee. Aside from these two samples and LAR 04316 (pieces) and Cumberland Falls (small aliquot in this study), the ε^{53}Cr values of the remaining aubrite samples, including those reported by Shukolyukov and Lugmair (2004), correlate with their respective Mn/Cr ratios and form a ^{53}Mn-^{53}Cr correlation line with a slope of 0.225 ± 0.046 and an intercept of ε^{53}Cr of 0.50 ± 0.16 (MSWD = 9.8, N = 8; Figure 4a), modeled by *Isoplot R* (Vermeesch, 2018), model 3 (MSWD >> 1). There is also a correlation between ε^{53}Cr values and δ^{53}Cr values (Figure 4b).

4. Discussion

4.1 ε^{54}Cr systematics and the number of enstatite achondrite parent bodies

One of the complications for precise Cr isotope measurements of meteorites is their susceptibility to cosmic ray induced modifications. In aubrites, which are mostly composed of Fe-poor enstatite, this effect is negligible because iron is the primary target for producing Cr isotopes via cosmic ray exposure (Shima and Honda, 1966). Aubrites also have short cosmic ray exposure ages [up to 100 Ma; (Eugster, 2003; Lorenzetti et al., 2003)] compared to other meteorites such as mesosiderites which do not exhibit any cosmogenic effects on Cr isotopes themselves (Eugster, 2003; Trinquier et al., 2007). As such, modification of the Cr isotope systematics of aubrites by such processes is unlikely and not further considered.

The combination of ε^{54}Cr and Δ^{17}O signatures has been applied to investigate the kinship of Solar System materials (Trinquier et al., 2007; Warren, 2011; Zhu et al., 2021b). Most aubrites for which these data exist have indistinguishable ε^{54}Cr and Δ^{17}O values (Figure 1b), supporting the idea that the majority of aubrites originated from a single parent body. In addition, aubrites also have indistinguishable O and Cr isotopic composition with respect to the Earth, Moon and enstatite chondrites (Figure 1a), and similar ε^{54}Cr values but different Δ^{17}O values compared to R chondrites (Bischoff et al., 2011; Zhu et al., 2021a). Aside from the main aubrite population, some isotopically

distinct samples exist: Shallowater, a chondritic clast within Cumberland Falls, and Khor Temiki.

Although Shallowater has an indistinguishable $\Delta^{17}O$ value (-0.02 ± 0.02) from other aubrites (Newton et al., 2000), the $\varepsilon^{54}Cr$ value reported here is lower than the ones of other aubrites and enstatite chondrites, suggesting a distinct parent body. Formation of Shallowater from a distinct parent body is also consistent with its higher Fe and Cr content (Rubin, 2015; Watters and Prinz, 1979), heavier Zn and Fe isotope compositions (Moynier et al., 2011a; Wang et al., 2014), higher HSE contents (van Acken et al., 2012a) and more complex cooling history (Keil et al., 1989) relative to other aubrites.

The distinct and variable $\Delta^{17}O$ values reported for Cumberland Falls (Clayton et al., 1984; Miura et al., 2007; Newton et al., 2000) have been interpreted as representing the composition of ordinary chondrite-like clasts (Barrat et al., 2016; Verkouteren and Lipschutz, 1983) in this breccia. As such, the $\Delta^{17}O$ composition of Cumberland Falls cannot be taken as a robust genetic signature. Nonetheless, the sample of Cumberland Falls analyzed here has an indistinguishable $\varepsilon^{54}Cr$ value from other aubrites (with the exception of Shallowater) and enstatite chondrites. This value differs significantly from that of ordinary chondrites [-0.39 ± 0.09, 2SD; (Pedersen et al., 2019; Qin et al., 2010; Trinquier et al., 2007)] and, thus, suggests that the sample analyzed here is not compromised by the presence of ordinary chondrite material. Given its indistinguishable $\varepsilon^{54}Cr$ value with other aubrites, Cumberland Falls appears to belong

to the main group-aubrites. However, oxygen isotopes indicates that some limited isotopic heterogeneity existed within this meteorite.

The aubrite Khor Temiki, is characterized by a ε^{54}Cr value of 0.18 ± 0.04 that is slightly higher but overlaps within uncertainties with some other aubrites, including Norton County (0.06 ± 0.09), LAR 04316 (0.06 ± 0.09) and Cumberland Falls (0.06 ± 0.09). Despite its slightly elevated ε^{54}Cr value, Khor Temiki has an indistinguishable O isotope composition (Δ^{17}O = 0.01 ± 0.02) from other aubrites (Barrat et al., 2016) and lacks other evidence that could be used to distinguish it from other aubrites. As such, based on ε^{54}Cr values, Khor Temiki may have originated from a distinct parent body than other aubrites or contain foreign, ^{54}Cr-rich clasts but additional data are required to test these hypotheses.

Itqiy (ε^{54}Cr = -0.27 ± 0.10) and MS-MU-19 (ε^{54}Cr = -0.25 ± 0.08) have the same ε^{54}Cr values, consistent with their petrological similarities: they are both enstatite-rich, they lack plagioclase and are depleted in Na, K and V (Harries and Bischoff, 2020; Patzer et al., 2001). Although Itqiy has similar O isotope compositions as ECs and aubrites (Patzer et al., 2002), both Itqiy and MS-MU-19 show clearly different ε^{54}Cr values from enstatite chondrites [0.03 ± 0.13; 2SD, N = 12; (Mougel et al., 2018; Qin et al., 2010; Trinquier et al., 2007)]. This observation suggests that they do not directly originate from enstatite chondrites and the aubrite (both main-group ones and Shallowater) parent bodies. The same ε^{54}Cr values of Itqiy and MS-MU-19, and the similar petrology of Itqiy, NWA 2526, MS-MU-19 and MS-MU-36, supports that there

may be at least an additional enstatite-dominated asteroid (probably with a size of 500 km; Harries and Bischoff, 2020) in the Solar System, i.e. the Itqiy parent body. Thus, combining petrological arguments with the ε^{54}Cr systematics allows us to distinguish at least three isotopically distinct reservoirs that generated enstatite-rich achondrites: the main-group aubrite parent body with ε^{54}Cr = 0.06 ± 0.12 (2SD, N = 7), Shallowater parent body with ε^{54}Cr = -0.12 ± 0.04 and probably, the Itqiy parent body with ε^{54}Cr = -0.26 ± 0.03 (2SD, N = 2). Note that this is not the same situation as for the ε^{54}Cr data for monomict ureilites, which shows a broad compositional spectrum (i.e. continuous variation, instead of showing three groups of enstatite achondrites), which is interpreted as planetary mantle heterogeneity in a single parent body (Zhu et al., 2020b).

Hence, among the enstatite achondrites, only the ε^{54}Cr values of main-group aubrites are indistinguishable from the Earth [(0.09 ± 0.12; 2SD, N = 17; (Mougel et al., 2018; Trinquier et al., 2007)] and the Moon [(0.09 ± 0.08; (Mougel et al., 2018)], and enstatite chondrites [0.02 ± 0.11; 2SD, N = 13; (Mougel et al., 2018; Qin et al., 2010; Trinquier et al., 2007; Zhu et al., 2021b)] and most enstatite chondrite chondrules: –0.11 ± 0.13 (2SD, N = 7) (Zhu et al., 2020a), which may indicate that they originated from a common precursor reservoir. However, the main-group aubrites cannot directly derive from enstatite chondrites, which will be further discussed in section 4.5. Finally, the ε^{54}Cr sequence for non-carbonaceous achondrites (including the planets and asteroids) can be updated as: Earth = Moon = main-group Aubrites > Shallowater > Itqiy ≈ Mars > Angrites > Winonaites ⩾ Acapulcoite-Lodranite Clan ⩾ Vesta >

Ureilites (Li et al., 2018; Mougel et al., 2018; Qin et al., 2010; Trinquier et al., 2007; Yamakawa et al., 2010; Zhu et al., 2019b; Zhu et al., 2020b).

4.2 Chromium stable isotope fractionation during sulfide mineral crystallization

All aubrites and Itqiy (EH7-an; bulk) are enriched in the heavier isotopes of Cr with respect to chondrites and other differentiated parent bodies (Fig. 3). These distinct stable isotopic compositions must be the result of parent body processes since chondrites from different groups have homogeneous δ^{53}Cr values (Bonnand et al., 2016b; Schoenberg et al., 2016; Zhu et al., 2021a), which excludes Cr stable isotope fractionation by nebular processes. Chromium isotopes are fractionated during planetary processes, such as partial melting and fractional crystallization as observed for the Earth (Bonnand et al., 2020b; Schoenberg et al., 2008; Shen et al., 2019), the Moon (Bonnand et al., 2016a; Sossi et al., 2018) and Vesta (Zhu et al., 2019c). Moreover, equilibrium isotope fractionation by evaporation processes between oxidized vapor and reduced residue has been documented for lunar and Howardite-Eucrite-Diogenite (HED) samples (Sossi et al., 2018; Zhu et al., 2019c).

Aubrites are dominated by silicate minerals [≈ 99%, except for Shallowater; (Watters and Prinz, 1979)], predominantly enstatite and a minor plagioclase, diopside and forsterite. However, these silicate minerals are usually poor in Cr, mostly < ~300 ppm (Watters and Prinz, 1979). Chromium abundances in metals, i.e., kamacite and schreibersite, are even lower, ~20 ppm (Casanova et al., 1993). On the contrary, sulfide

minerals are usually rich in Cr, e.g., troilite, daubreelite, alabandite, caswellsilverite and heideite can contain between ~0.5 (troilite) to ~37.4 (daubreelite) wt% Cr. The much higher Cr content of sulfides relative to that of silicates and high silicate abundance in main-group aubrites indicate that the bulk contents of Cr are controlled primarily by silicate and sulfide minerals, since metal does not contribute much to the Cr budget (Casanova et al., 1993). Therefore, the variations of the Cr contents and of the δ^{53}Cr values possibly result from a two-endmember mixing between isotopically heavy and Cr-poor silicates and isotopically light and Cr-rich sulfides. This is consistent with *ab initio* calculations that predict that CrS and daubréelite ($FeCr_2S_4$) are isotopically light compared to silicates (Moynier et al., 2011b). However, there is no correlation between [δ^{53}Cr] and [1/Cr] (figure 2c) as would be expected for simple mixing between sulfides (high Cr content and low δ^{53}Cr values) and silicates (low Cr content and high δ^{53}Cr values). This could be due to secondary processes (i.e. an impact event) that result in the equilibration of the Cr isotopes at the meteorite scale, while the heterogeneous distribution of Cr-rich sulfide would significantly affect the Cr contents of the different chips of a same meteorite. For example, Cumberland Falls shows variable Cr contents, 945 ppm in this study and 549 ppm in Shukolyukov and Lugmair (2004). Thus, it is difficult to determine the representative Cr contents (also the Mn/Cr ratios) in bulk aubrite samples (i.e., different chips from one aubrite can have similar Cr stable isotope compositions but distinct Cr contents), if the sample size is not large

enough, e.g., < 600 mg (Stracke et al., 2012). This sample heterogeneity effect is also discussed in section 4.4.

This two-endmember (silicate-sulfide) mixing for aubrites is reminiscent of the positive correlation between Fe content and Fe isotopes in aubrites (Wang et al., 2014). This has been interpreted as the mixing of isotopically light Fe from sulfide-rich metal with isotopically heavy, Fe-poor silicates (mantle material). In detail, Wang et al. (2014) argued the silicate in aubrites represent the mantle of aubrite parent body, while the metal is likely some residue in the mantle from incomplete metal-silicate differentiation (Casanova et al., 1993). A common origin for the δ^{53}Cr and δ^{56}Fe variability of aubrites, ignoring the anomalous Shallowater, is supported by their correlated variability (Figure 2d). However, because of its low Cr abundances, the Cr-poor Fe-Ni metal in aubrites (Casanova et al., 1993) cannot account for the Cr isotope variations. Instead, it is more likely that the variable presence of sulfide minerals (e.g., troilite) that are both rich in Fe and Cr (Keil, 2010a; Watters and Prinz, 1979) caused the observed correlated effects between Cr and Fe isotopes. This interpretation is also supported by experimental data that show equilibrium Fe isotope fractionation increases with sulfur content to ~0.4 ‰ at 18 wt.% sulfur in metal coexisting with silicate melt (Shahar et al., 2015).

4.3 Isotopically heavy Cr in the mantle of aubrite parent bodies induced by core formation

In order to use Cr stable isotopes to understand the formation and differentiation of the aubrite parent bodies (main-group and Shallowater), it is necessary to estimate

the δ^{53}Cr composition of their mantles. The depletion of HSE (van Acken et al., 2012a; van Acken et al., 2012b) in aubrites compared to chondrites is evidence that their parent body underwent core formation. However, given the high Cr content of sulfides compared to silicates and their heterogeneous distribution within aubrites, obtaining a pure silicate mantle composition requires a sulfide-free sample. Also, troilite, as the major sulfide minerals in aubrites, contains Fe (~60%; (Watters and Prinz, 1979), which given their chemical composition are usually interpreted as reflecting leftover core material after incomplete core-mantle differentiation of the aubrite parent body (Casanova et al., 1993). As explained above, Norton County has the highest Mg# (99.9) and a bulk Cr content (180 ppm) comparable to the Cr content of typical silicate minerals [<0.04%, (Keil, 2010a; Watters and Prinz, 1979)]. As such, the Cr isotope composition of Norton County, δ^{53}Cr = 0.24 ± 0.03 ‰, is taken as the best estimation of the mantle Cr isotope composition. Considering the silicate-sulfide mixing model for Cr stable isotope variation, Bustee with heavier δ^{53}Cr value（0.30 ± 0.03 ‰) may also indicate the silicate compositions and bulk silicate main-group aubrite parent body. The elevated Cr contents and lower Mg# for Bustee can be also caused by sample heterogeneity. However, the δ^{53}Cr values for both Norton County and Bustee are resolvably heavier than those of chondrites, Shallowater and Itqiy. Given that Shallowater and Itqiy are unique samples, their bulk δ^{53}Cr values of 0.10 ± 0.03 ‰ and -0.03 ± 0.03 ‰ are our best approximation for their parent bodies' mantle. The mantle δ^{53}Cr difference between the three parent bodies (Figure 3), consistent with their ε^{54}Cr

signatures (Figure 1b and 2a), confirms the existence of at least three enstatite achondrite parent bodies in the Solar System. This is also consistent with the difference in the mantle Mg# between three parent bodies (Figure 2b).

Chondrites have homogeneous Cr stable isotope compositions, with δ^{53}Cr = -0.12 ± 0.04 ‰ (2SD, N = 42) (Bonnand et al., 2016b; Schoenberg et al., 2016; Zhu et al., 2021a). The δ^{53}Cr values of all aubrites, including main-group, Shallowater and Itqiy are heavier than bulk chondrites as well as the mantles of other planetary bodies: Earth [-0.12 ± 0.04 ‰; (Jerram et al., 2020; Schoenberg et al., 2008; Sossi et al., 2018)], Moon [-0.21 ± 0.03 ‰; (Bonnand et al., 2016a; Sossi et al., 2018)] and Vesta [-0.22 ± 0.03 ‰; (Zhu et al., 2019c)] (Figure 3). These isotopic offsets: $\Delta_{\text{main-group aubrites-Chondrites}}$ = 0.36 ± 0.05 ‰, $\Delta_{\text{Shallowater-Chondrites}}$ = 0.22 ± 0.05 ‰ and $\Delta_{\text{Itqiy-Chondrites}}$ = 0.09 ± 0.05 ‰, combined with significant depletion in Cr between aubrites and chondrites (including enstatite chondrites) may provide significant insights into the origin of the bulk silicate aubrite parent bodies.

Several mechanisms may have caused the Cr stable isotope difference between chondrites and the bulk silicate aubrite parent bodies: 1) igneous processes, such as partial melting (e.g., Zhu et al., 2019c), 2) volatilization processes (e.g., Sossi et al., 2018; Zhu et al., 2019c) or 3) core formation (e.g., Bonnand et al., 2016b; Moynier et al., 2011b). Partial melting is a possible way to fractionate Cr stable isotopes, because of the potential exchange between "heavy" Cr^{3+} and "light" Cr^{2+} in residue and melts (e.g., Shen et al., 2019; Zhu et al., 2019c). This is also consistent with the fact that the

aubrites are depleted in basaltic components (Wilson and Keil, 1991). However, given the highly reduced conditions during aubrite differentiation (IW-2—IW-6), both the melt and residue are dominated by Cr^{2+} (Bell et al., 2020; Berry et al., 2006), and therefore partial melting is unlikely to fractionate Cr stable isotopes. Moreover, for the Earth's mantle, which is more oxidized than aubrites and contains both Cr^{2+} and Cr^{3+}, the extent of Cr isotopic fractionation is less than ~0.15 ‰ (based on the $\delta^{53}Cr$ difference between terrestrial chromite-free peridotites and chromites (Shen et al., 2015). This difference is much lower that what would be required to explain the difference between chondrites and main-group aubrite parent body (0.36 ± 0.05 ‰). Hence, partial melting cannot result in the observed Cr stable isotopic variations of the aubrites.

The volatility of Cr decreases together with fO_2, e.g., at fO_2 below IW = -2 and atmospheric pressure Cr becomes more refractory than Mg (Sossi et al., 2019). Thus, under reduced conditions volatile loss of Cr from aubrites should have resulted in concomitant losses of Mg which is not observed (Sedaghatpour and Teng, 2016). However, it is unknown whether the sulfur content affect the volatility of Cr, so we cannot totally exclude that the isotopically heavy Cr in aubrites is the result of evaporation (e.g., if sulfur increase the Cr volatility), considering that aubrite sulfides are rich in Cr (Watters and Prinz, 1979). Given the distinct Cr content and metal-sulfide abundance between bulk silicate aubrite parent body and ECs (Keil, 1968), and that Cr is not likely to be lost by volatilization, stable isotope fractionation

associated with core formation may be the most plausible mechanism to account for the heavier Cr in aubrites compared to chondrites.

The size of the aubrite parent body is not known, but is hypothesized to be probably <100 km based on models of the solubility of volatiles in basaltic melts of the aubrite parent body (Wilson and Keil, 1991). Chromium is mostly chalcophile, not siderophile, under the prevalent very low fO_2 of enstatite meteorites and low pressure conditions (Wood et al., 2008) and, thus, there should also be little Cr dissolved in the metallic core of the aubrite parent bodies. However, the partition coefficient of Cr into metal increases with the increasing sulfur content (Wood et al., 2014), which is also consistent with the high Cr content in the sulfide minerals in aubrites (Keil, 2010a; Watters and Prinz, 1979). Previous experiments have predicted that there should be very limited Cr stable isotope fractionation during metal-silicate segregation (Bonnand et al., 2016b). However, the experiments did not consider the composition effect of S, so it does not necessarily apply to aubrites, because the extremely reduced conditions during differentiation of aubrite parent body would likely form a core with abundant S. At present, it is difficult to provide a quantitative model to test the hypothesis that Cr isotope fractionation occurred during core formation. The core of the aubrite parent body may have contained both Fe-Ni-Si metal and sulfide. Unfortunately, even if the Cr contents in metal and sulfide were known, the proportions of metal and sulfide in the core cannot be estimated reliably, and thereby, the Cr contents in the core remain unknown.

The three enstatite achondrite parent bodies possess higher δ^{53}Cr values relative to other planets/asteroids, including Earth, Moon and Vesta (Figure 3). Earth possesses similar δ^{53}Cr values as chondrites (Bonnand et al., 2016b; Jerram et al., 2020; Schoenberg et al., 2016; Schoenberg et al., 2008; Sossi et al., 2018), suggesting that core formation did not fractionate Cr stable isotopes, which is consistent with the high temperature of Earth's core formation and further confirmed by high-pressure and high-temperature experiments (Bonnand et al., 2016b). Due to the relatively low temperature of the lunar core-mantle boundary (Cr is only siderophile at high temperature; (Corgne et al., 2008), only ~1% of the Moon's Cr would enter its core (Sossi et al., 2018). Therefore, the isotopically light Cr in lunar rocks is not due to core formation, but rather to a volatile loss during the magma ocean stage (Sossi et al., 2018). Also, Vesta, with a Cr-poor core, has lower δ^{53}Cr values than chondrites and enstatite achondrites, and its isotopically light Cr is interpreted to be caused by low-temperature volatile process rather than core formation (Zhu et al., 2019c). The Cr stable isotope composition of the Martian mantle is not well constrained, and in the future the physical conditions for the formation of the Martian core can be potentially traced with Cr isotopes.

4.4 Timescale of evolution of the main-group aubrite parent body

Aubrites are the product of magmatic differentiation on their parent body, which can potentially be dated using the ^{53}Mn-^{53}Cr chronometer (e.g., Shukolyukov and Lugmair, 2004). The ^{55}Mn/^{52}Cr ratio of Cumberland Falls reported by Shukolyukov

and Lugmair (2004), corresponding to 1.99, is different from the one determined here (8.40). However, the two portions of Cumberland Falls have the same ε^{53}Cr values. A similar observation has been made for leachates with variable ^{55}Mn/^{52}Cr ratios (0.04 – 9.58) for Bishopville (Shukolyukov and Lugmair, 2004) and two portions of LAR 04316 in this study, with ^{55}Mn/^{52}Cr ratios of 4.55 and 7.46, that all return similar ε^{53}Cr values, suggesting that internal Mn/Cr redistribution post-dates ^{53}Mn decay and lack of internal ^{53}Mn-^{53}Cr isochrons in aubrites. This situation does not mimic the leachates or minerals (with variable ^{55}Mn/^{52}Cr ratios) in some individual angrites (Glavin et al., 2004; Lugmair and Shukolyukov, 1998), eucrites (Lugmair and Shukolyukov, 1998) and ureilites (Yamakawa et al., 2010), which show variable ε^{53}Cr values. Considering the half-life of ^{53}Mn [(3.7 Myrs; (Holden, 1990)] combined with the Mn/Cr variability in LAR 04316, Cumberland Falls and Bishopville and the precision of the ε^{53}Cr data, the redistribution of Mn in these aubrites likely occurred after the extinction of ^{53}Mn, i.e. >20 Myrs (eight half-life of ^{53}Mn; Figure 5) after Solar System formation. Metamorphic redistribution of Mn and Cr on the meteorite scale might have occurred by impact processes and is also supported by the fractionated trace elements and by Ar-Ar ages (Biswas et al., 1980; Bogard et al., 2010). On the other hand, the heterogeneous distribution of Mn/Cr ratios in aubrites, mainly caused by Mn- or Cr-rich sulfide minerals, suggest that it will be difficult to determine the true ^{55}Mn/^{52}Cr ratios of bulk aubrites. In other words, the ε^{53}Cr of each meteorite sample could be representative of the bulk meteorite, while the ^{55}Mn/^{52}Cr ratio may not be, since

different sample fractions with variable Mn/Cr ratios have the same ε^{53}Cr values.

Aubrites do not all fall on the same line in a ε^{53}Cr against Mn/Cr ratio diagram (Figure 4a), including the three data points from Shukolyukov and Lugmair (2004). To limit nugget effects of Cr-rich sulfides in aubrites (that would result in a variation of Mn/Cr ratios of bulk aubrites and would prohibit obtaining the "true" Mn/Cr ratios of bulk aubrites), we focus on the largest samples that should be more representative of the bulk sample composition. For this reason, we only consider the largest LAR 04316 sample (Table 1). Similarly, for Cumberland Falls, we use the data reported by Shukolyukov and Lugmair (2004) (Cumberland Falls-SL; Table 1) that is obtained from a large sample (905 mg) compared to ours (42 mg). Excluding also Bustee, which has a very different Mn/Cr ratio in our study (1.42) compared to the literature (6.08, Watters and Prinz, 1970) and Shallowater that is likely originating from a different parent body, we obtain a correlation line (Figure 4a) for the main-group aubrites. Using a model 3 (due to MSWD >> 1) regression of *Isoplot R*, this ^{53}Mn-^{53}Cr correlation line for the main-group aubrites has an intercept (i.e., initial ε^{53}Cr value) of 0.50 ± 0.16 and a slope of 0.225 ± 0.046 (MSWD = 9.8, N = 8). This slope corresponding to a ^{53}Mn/^{55}Mn ratio of $(2.55 \pm 0.52) \times 10^{-6}$) can be translated to an absolute date of 4562.5 ± 1.1 Ma, anchored to the U isotope corrected U-Pb age of D'Orbigny angrite (Amelin, 2008; Brennecka and Wadhwa, 2012; Glavin et al., 2004). This date is similar to that reported in Shukolyukov and Lugmair (2004), but in view of our new stable isotope data, we interpret it was controlled by mixing of silicate and

sulfide minerals, and the time significance needs to be discussed.

Most aubrites are breccias (Keil, 2010a) and, as such, many are mixtures of rocks with a different history. Hence, these rocks do not a have simple igneous history, which limits the use of the ^{53}Mn-^{53}Cr system to obtain reliable age information such as previously done for eucrites and diogenites (Trinquier et al., 2008b) and angrites (Zhu et al., 2019b). This mixing history is also supported by the stable Cr isotope data (section 3.2 and 3.3), suggesting that the δ^{53}Cr variation in aubrites is mainly controlled by the relative abundance of sulfides and silicates. Finally, the δ^{53}Cr values of aubrites positively correlate with their ε^{53}Cr values (Figure 4b). Their ε^{53}Cr values mainly reflect the true ^{55}Mn/^{52}Cr ratios, because the Mn/Cr heterogeneity and secondary processes for aubrites make it difficult to determine the representative Mn/Cr ratios for bulk aubrite samples. This correlation indicates that the Cr in aubrites are mixtures of Cr-rich sulfide (low Mn/Cr, low ε^{53}Cr and low δ^{53}Cr) and Cr-poor silicates (high Mn/Cr, high ε^{53}Cr and high δ^{53}Cr).

Since aubrites are igneous rocks that experienced magmatic processes, both silicate and sulfide should crystallize from the melt and derive from a common initial reservoir. Considering this, the conditions for silicate and sulfide to define an isochron are: 1) to be formed at the same time (crystallization from the same magma), 2) to originate from the same reservoir (the melt) and 3) that possible later impact metamorphism would not change the Mn/Cr ratios and Cr isotope compositions of the bulk aubrites (closed system; but this could reset and flatted the internal isochron for

individual aubrites). In this respect, an external ^{53}Mn-^{53}Cr isochron age for main-group aubrites of 4562.5 ± 1.1 Ma would be valid and with time significance.

As suggested before for Mn-Cr whole rock isochrons of other planetary bodies (Lugmair and Shukolyukov, 1998; Trinquier et al., 2008b; Zhu et al., 2019b; Zhu et al., 2020b), the age from the bulk aubrite isochron likely records the timing of the differentiation of the main-group aubrite parent body, similar to those of Vesta, the angrite and ureilite parent bodies (Trinquier et al., 2008b; Zhu et al., 2019b; Zhu et al., 2020b). Mineral crystallization would also fractionate Mn/Cr ratios between aubrites. Enstatite in aubrites is typically characterized by low Mn and Cr contents (< 400 ppm for Mn and < 200 ppm for Cr) (Watters and Prinz, 1979). As such, the Mn/Cr fractionation between different main-group aubrites reflects variable incorporation of Mn-rich and Cr-rich minerals (e.g. oldhamite, daubreelite, caswellsilverite, heideite and niningerite) under reduced conditions during their crystallization. This age is very similar to the rock-forming age of Shallowater 4562.4 ± 0.2 Ma (Pravdivtseva et al., 2017), suggesting that the magmatic process for the enstatite achondrite parent bodies occurred at the same time. On the other hand, the differentiation time of the enstatite achondrite parent bodies is delayed compared to the ureilite parent body with ^{53}Mn-^{53}Cr age of 4566.7 ± 1.5 Ma (Zhu et al., 2020b), Vesta with ^{53}Mn-^{53}Cr age of 4564.8 ± 0.6 Ma (Trinquier et al., 2008b) and the angrite parent body with ^{53}Mn-^{53}Cr age of 4563.2 ± 0.3 Ma (Zhu et al., 2019b) when anchored to D'Orbigny.

4.5 The relationship between enstatite chondrites and aubrites and core formation based on the initial ε^{53}Cr values

The initial ε^{53}Cr value of the aubrite precursors defined by the intercept of the Mn-Cr isochron with the ordinate axis provides additional insight into the origin of aubrites and their potential genetic relationship to enstatite chondrites. The initial ε^{53}Cr value for the main-group aubrites (0.50 ± 0.16, 2σ) is significantly higher than the average ε^{53}Cr value of bulk enstatite (0.16 ± 0.06, 2SD, N = 12), Rumuruti (0.23 ± 0.05, 2SD, N = 12), carbonaceous (-0.04 ~ 0.27) and ordinary (0.19 ± 0.10, 2SD, N = 23) chondrites (Mougel et al., 2018; Qin et al., 2010; Trinquier et al., 2008b; Zhu et al., 2021a; Zhu et al., 2021b). This high initial ε^{53}Cr value is also consistent with the intercept of the three-point isochron from Shukolyukov and Lugmair (2004), but its implications have never been fully discussed. Moreover, whole-rock Mn-Cr isochrons for Vesta, angrite and ureilite parent bodies, which record Mn-Cr ages of ~2.5 Ma, ~4.1 Ma and ~0.5 Ma after CAIs, respectively, record lower initial ε^{53}Cr values than their parental melts of -0.12 ± 0.05 (Trinquier et al., 2008b), -0.10 ± 0.06 (Zhu et al., 2019b) and -0.24 ± 0.10 (Zhu et al., 2020b). It should also be noted that the initial ε^{53}Cr value of main-group aubrites is much higher than that of EC chondrules, -0.13 ± 0.07 (Zhu et al., 2020a), which are potential building blocks of enstatite achondrite parent bodies (e.g., Johansen et al., 2015). The higher initial ε^{53}Cr value as recorded by the aubrite whole rock isochron implies that the parent magma of the aubrites had a supra-chondritic Mn/Cr ratio. Shallowater is characterized by a low ε^{53}Cr value (0.00 ± 0.02) compared

to other aubrites, indicating that it evolved from another source with a resolvably lower initial ε^{53}Cr value than those of the main-group aubrites. Combined with the ε^{54}Cr systematics, the Cr isotope evidence strongly suggests that the main-group and Shallowater aubrites should have distinct genetic origins.

The elevated initial ε^{53}Cr value of main-group aubrites compared to that of other asteroids (e.g., Vesta, angrite and ureilite parent bodies) in the Solar System, indicates that their parent melt had a high, non-chondritic, Mn/Cr ratio. A supra-chondritic Mn/Cr ratio of the parent magma of main-group aubrite parent body can occur either if the aubrite parent body precursor was characterized by a Mn/Cr ratio higher than chondrites or if the Mn/Cr ratio of primitive melt for aubrites changed the during the early evolution of the parent body. Note that the partition coefficient between FeS and silicate melt, i.e. $D^{FeS/silicate\ melt}$, of Cr is higher than that of Mn (Berthet et al., 2009), so the S-rich core formation process can efficiently increase the Mn/Cr ratio and ε^{53}Cr value of the silicate melt of main-group aubrite parent body. This scenario is supported by the S-rich core formation triggering large Cr stable isotope fractionation of aubrites (see section 4.3). This is also consistent with that other S-poor and early formed rocky planets, i.e. Vesta, angrite and ureilite parent body, have sub-chondritic initial ε^{53}Cr values (Trinquier et al., 2008b; Zhu et al., 2019b; Zhu et al., 2020b).

Given the otherwise close kinship between aubrites and enstatite chondrites expressed by their similarly reduced conditions, mineralogy and isotope compositions (including Δ^{17}O, ε^{54}Cr and ε^{50}Ti values), it is more likely that the initial Mn/Cr ratio of

the aubrite parent body was close to chondritic. Therefore, aubrites may be the product of post-core formation melting on an enstatite chondrite-like parent body that accreted (Zhu et al., 2020a) and melted very early (e.g., < 2 Myr after Solar System formation) to generate ^{53}Cr ingrowth prior to aubrite formation (Figure 5). Such early melting is typically associated with large scale differentiation on other asteroids such as Vesta (Schiller et al., 2011; Trinquier et al., 2008b) or the angrite parent body (Schiller et al., 2015; Zhu et al., 2019b) due to the abundance of ^{26}Al at this time.

5. Conclusions

This work reported both the mass-independent and mass-dependent Cr isotope data for bulk enstatite achondrites, providing insights into the formation of their parent bodies in three ways, nucleosynthetic (ε^{54}Cr; origin), stable (δ^{53}Cr; process) and radiogenic (ε^{53}Cr; timing) isotope fractionations:

1. The ε^{54}Cr systematics define three groups of enstatite achondrites that likely represent three parent bodies, the main-group aubrite parent body with ε^{54}Cr of 0.06 ± 0.12 (2SD), Shallowater parent body with ε^{54}Cr = -0.12 ± 0.04 and Itqiy parent body with ε^{54}Cr = -0.26 ± 0.03 (2SD, N =2). This is consistent with their different δ^{53}Cr compositions: 0.24 ± 0.03 ‰, 0.10 ± 0.03 ‰ and -0.03 ± 0.03 ‰, respectively.

2. Mixing of different proportions of sulfides and silicates possibly result in the Cr stable isotope variation in aubrites. The globally heavy Cr in aubrites relative to that in chondrites could reflect large-scale Cr stable isotope equilibrium fractionation between sulfide (light) and silicate (heavy) followed by segregation of sulfide during core formation, resulting in isotopically heavy mantle.

3. The aubrite samples record heterogeneous distribution of Mn and Cr. The ^{53}Mn-^{53}Cr correlation for the main-group aubrites (except Bustee) is a mixing line but also an isochron that holds a time significance of the crystallization of silicate and sulfide minerals and return an absolute age of 4562.5 ± 1.1 Ma (i.e. 4.8 ± 1.1 Ma after CAIs). This most likely represents the age of the silicate differentiation on the

main-group aubrite body. The absence of internal Mn-Cr isochrons for individual aubrites implies that they underwent metamorphic redistribution of Mn and Cr after ^{53}Mn extinction, maybe following a disruption event of the parent body.

4. The initial ε^{53}Cr value for main-group aubrites are higher than the ε^{53}Cr values for enstatite chondrites, suggesting the main-group aubrites did not directly originate from enstatite chondrites. Instead, the high initial ε^{53}Cr values of main-group aubrites are potentially caused by an early sulfur-rich core formation that efficiently increased the Mn/Cr ratio of the silicate fraction.

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Appendix

Individual data for TIMS total-evaporation measurements

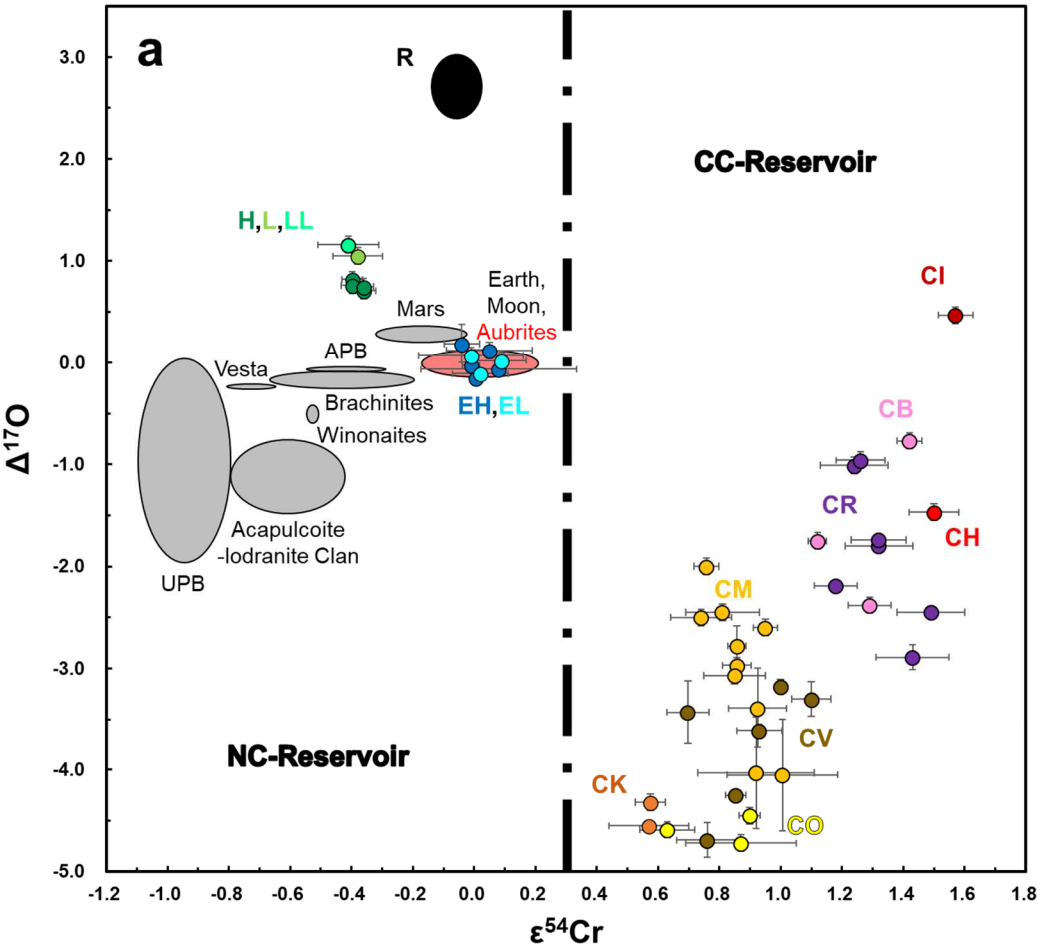
Sample	Itqiy		MS-MU-19		Allende		DTS-1	
N	**ε^{53}Cr**	**ε^{54}Cr**	**ε^{53}Cr**	**ε^{54}Cr**	**ε^{53}Cr**	**ε^{54}Cr**	**ε^{53}Cr**	**ε^{54}Cr**
1	0.50	-0.33	1.87	-0.15	0.19	1.01	0.19	0.31
2	0.56	0.06	1.78	-0.26	0.11	0.75	0.11	0.30
3	0.60	-0.23	1.78	-0.10	0.11	0.90	0.05	0.18
4	0.48	-0.55	1.71	-0.50	-0.04	0.67	0.05	0.41
5	0.44	-0.59	1.87	-0.29	0.12	1.01	-0.04	0.05
6	0.55	-0.22	1.65	-0.45	0.08	0.82	-0.01	0.08
7	0.43	-0.44	1.76	-0.33	0.05	0.84	-0.01	0.10
8	0.65	-0.20	1.72	-0.11	0.02	0.72	0.05	0.22
9	0.55	-0.16	1.77	-0.29	0.03	0.85	-0.10	-0.11
10	0.59	-0.17	1.66	-0.15	0.01	0.55	0.11	0.28
11	0.52	-0.07	1.80	-0.15			-0.04	0.01
12	0.46	-0.32					0.00	0.01
13	0.50	-0.21						
14								
Average	**0.52**	**-0.27**	**1.76**	**-0.25**	**0.07**	**0.81**	**0.03**	**0.15**
2SD	0.13	0.37	0.14	0.27	0.13	0.29	0.16	0.31
2SE	0.04	0.10	0.04	0.08	0.04	0.09	0.05	0.09

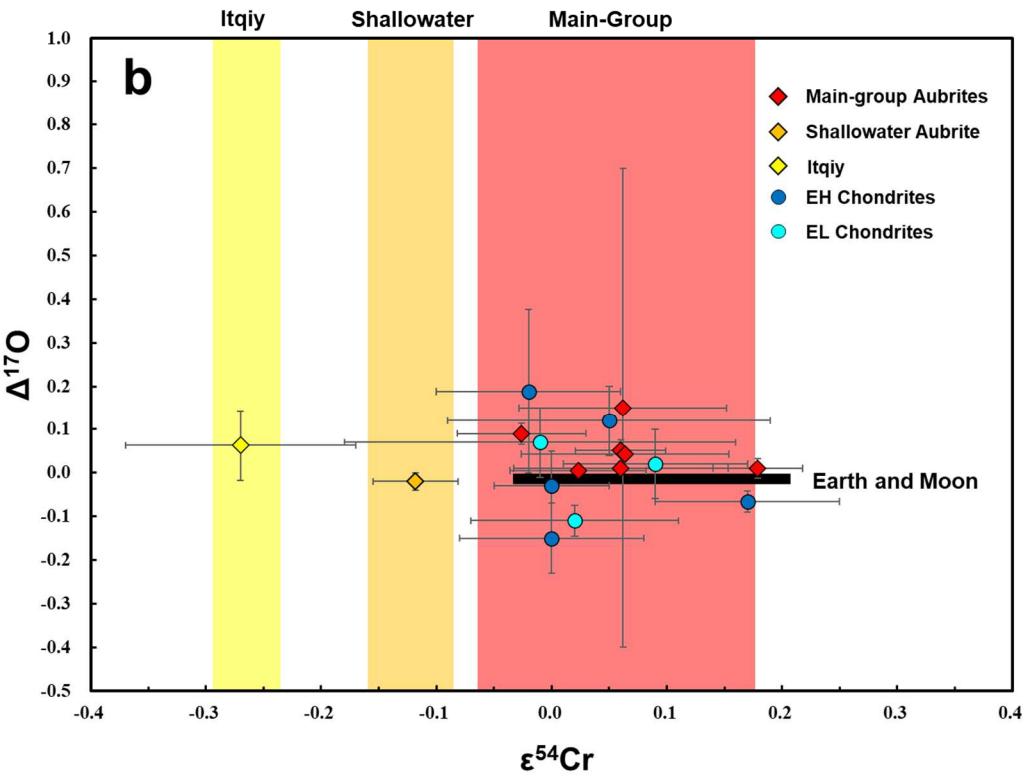
Table 1 Cr (both mass-independent and mass-dependent) and O isotopes data for aubrites.

Name	Fall/Find	Texture/Type	Mass (mg)	Cr content (ppm)	Mg#	Fe/Cr	$^{55}Mn/^{52}Cr$	$\varepsilon^{53}Cr$	2SE	$\varepsilon^{54}Cr$	2SE	N/Ref.	$\Delta^{17}O$	Error	Ref.	$\delta^{53}Cr$	2SD	N
Khor Temiki	Fall	Fragmental breccia	39.8	742	98.8	14.6	3.77	**1.39**	0.01	**0.18**	0.04	5	0.01	0.02	[2]	**0.22**	0.03	2
Bustee	Fall	Regolith breccia	33.7	2221	99.1	3.5	1.42	**2.41**	0.03	**0.02**	0.06	5	0.01	0.00	[2]	**0.30**	0.03	2
Norton County	Fall	Fragmental breccia	40.0	180	99.9	6.5	4.37	**1.53**	0.06	**0.06**	0.09	5	0.01	0.01	[2]	**0.24**	0.03	1
ALH 78113	Find	Fragmental breccia	43.5	1439	98.1	11.9	2.58	**0.97**	0.02	**-0.03**	0.06	5	0.09	0.02	[3]	**0.12**	0.03	2
ALH 84007	Find	Fragmental breccia	50.7	758	99.2	9.1	2.34	**1.08**	0.01	**0.06**	0.04	5	0.05	0.03	[3]	**0.23**	0.03	2
LAR 04316 (powder)	Find	Fragmental breccia	46.0	1109	98.7	10.0	4.55	**1.48**	0.02	**0.06**	0.09	5	0.04	0.01	[2]	**0.23**	0.03	2
LAR 04316 (pieces)			26.4	861	98.7	13.1	7.46	**1.49**	0.00*	**0.06**	0.05	5				**0.16**	0.03	2
Shallowater	Find	Unbrecciated	30.9	4597	77.7	42.2	0.33	**0.00**	0.02	**-0.12**	0.04	5	-0.02	0.02	[3]	**0.10**	0.03	2
Peña Blanca Spring	Fall	Fragmental breccia	898	394			3.01	**1.16**	0.05			15/[1]	0.01	0.01	[2]			
Bishopville	Fall	Fragmental breccia	911	273			3.79	**1.34**	0.08			17/[1]	-0.05	0.06	[3]			
Cumberland Falls	Fall	Fragmental breccia	42.0	945	98.2	17.2	8.40	**1.11**	0.05	**0.06**	0.09	5	0.15	0.55	[3,4,5]	**0.19**	0.03	2
Cumberland Falls-SL			905	549			1.99	**1.00**	0.09			14/[1]						
Chondritic Clast in Cumberland Falls		Clast											0.65	0.02	[2]			
Itqiy#	Find	Unbrecciated-EH7		830	65.1	25.7		**0.52**	0.04	**-0.27**	0.10	13	0.06	0.08	[6]	**-0.03**	0.03	3
MS-MU-19#	Fall	Clast						**1.76**	0.04	**-0.25**	0.08	11						
Allende#	Fall	CV chondrite						**0.07**	0.04	**0.81**	0.09	10						
DTS-1#		Terrestrial dunite						**0.03**	0.05	**0.15**	0.09	12						

Note: References: [1] (Shukolyukov and Lugmair, 2004), [2] (Barrat et al., 2016), [3] (Newton et al., 2000), [4] (Clayton et al., 1984), [5] (Miura et al., 2007) and [6] (Patzer et al., 2002). The $\delta^{53}Cr$ and elemental data for Itqiy are from a bulk sample, while the $\varepsilon^{53}Cr$ and $\varepsilon^{54}Cr$ data for Itqiy and MS-MU-19 are from leached

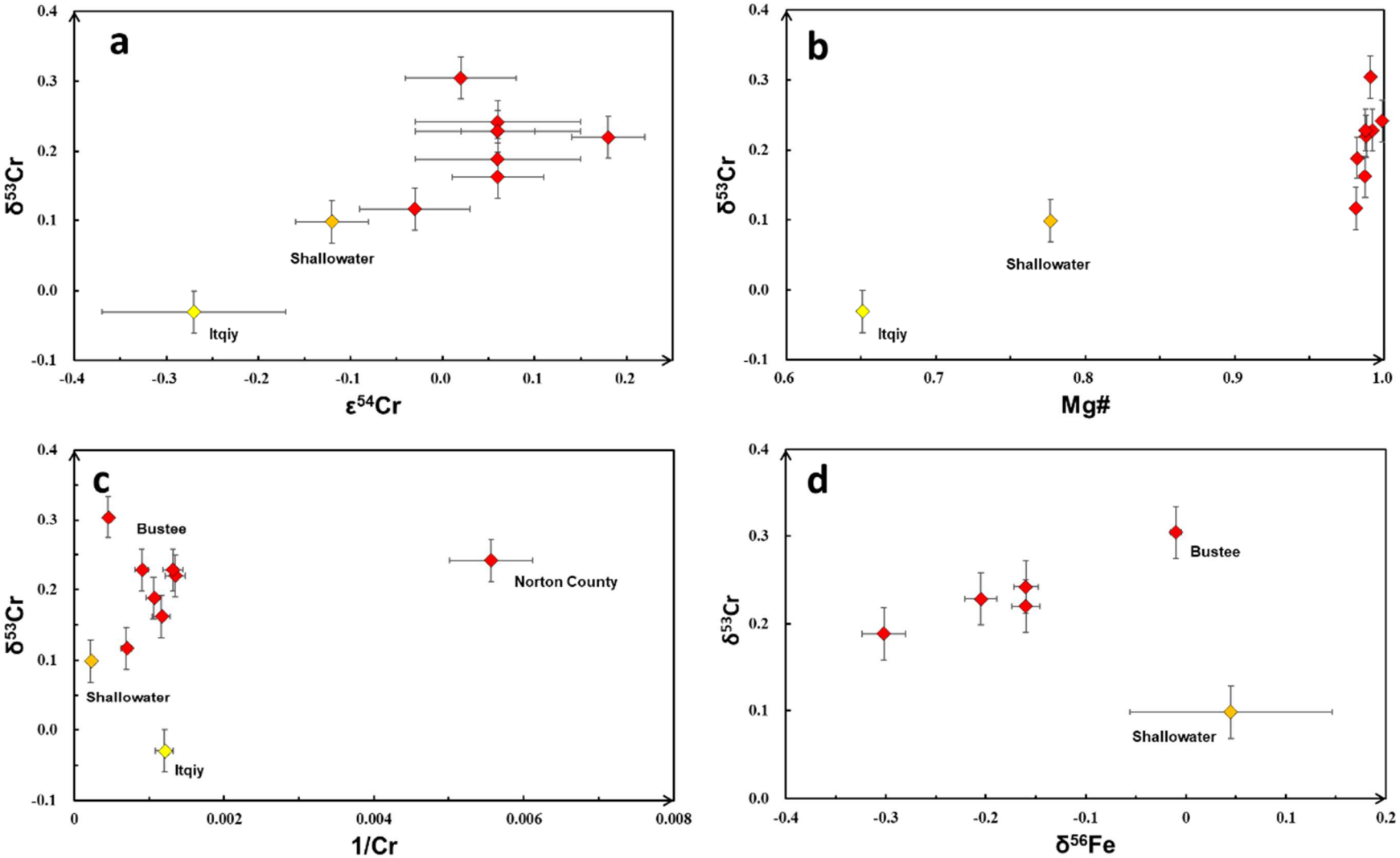
samples with mostly silicate (the metal and sulfide were removed the by HCl). The Cr content, Mg# (Mg/(Mg+Fe)×100%, atom ratios) and Fe/Cr (atom ratios) are measured by ICP-MS, with an error of 5-10% (2se), except for the data for bulk Itqiy that are from (Patzer et al., 2001). N is then number of runs. The data marked * indicates the 2SE uncertainty is less than 0.004. The data for sample marked # are measured by total evaporation method on Triton TIMS housed at Freie Universität Berlin.


a
R
CC-Reservoir
H,L,LL
Earth,
Moon,
Aubrites
Mars
APB
Vesta
Brachinites
EH,EL
Winonaites
Acapulcoite
-lodranite Clan
UPB
NC-Reservoir
CI
CB
CR
CH
CM
CV
CK
CO
Δ17O
ε54Cr


Itqiy
Shallowater
Main-Group
b
Main-group Aubrites
Shallowater Aubrite
Itqiy
EH Chondrites
EL Chondrites
Earth and Moon
Δ17O
ε54Cr

1

Figure 1a. The ε^{54}Cr - Δ^{17}O diagram for aubrites, enstatite chondrites and other Solar System materials. The area of enstatite meteorites (in red) overlaps with that of Earth-Moon system. The template and dataset (all the data cited are from published literature and no conference abstracts) are from Zhu et al. (2021b), and the data sources are mainly from Schiller et al. (2014), Trinquier et al. (2007), Yamashita et al. (2010), Qin et al. (2010), Göpel et al. (2015), Pedersen et al. (2019), Mougel et al. (2018), Clayton and Mayeda (1999), Clayton and Mayeda (1984), Greenwood et al. (2010), Clayton et al. (1991), Clayton et al. (1984), Greenwood et al. (2017), Newton et al. (2000), Schrader et al. (2011), Petitat et al. (2011), Zhu et al. (2020b), Zhu et al. (2020a), Zhu et al. (2019b), Li et al. (2018), Zhu et al. (2020b), Yamakawa et al. (2010), Greenwood et al. (2017), Zhu et al. (2020b), Ireland et al. (2020), Zhu et al. (2021a) and Bischoff et al. (2011). The colored circles are chondrites, while the grey shade areas indicate planets/asteroids (i.e., achondrite parent bodies). **Figure 1b.** The zoomed figure for the aubrite area of figure 1a. The enstatite achondrites are diamonds, and the error for ε^{54}Cr data are reported as the 2SE, while the error for grouping ε^{54}Cr (shades) of main-group aubrites and Itqiy is the 2SD. The ε^{54}Cr and Δ^{17}O for main-group aubrites overlap those the enstatite chondrite and Earth-Moon system (black bar), but Shallowater has a different ε^{54}Cr value compared to other aubrites, enstatite chondrites and Earth-Moon system. The red-orange-yellow shades indicate the three enstatite achondrite parent bodies inferred from ε^{54}Cr systematics.


a
b
c
d
Shallowater
Itqiy
Bustee
Norton County
δ53Cr
ε54Cr
Mg#
1/Cr
δ56Fe

21

Figure 2a. ε^{54}Cr-δ^{53}Cr values for enstatite achondrites, with the red diamonds for main-group aubrites, the orange diamond for Shallowater and the yellow diamond for Itqiy. There is no correlation between ε^{54}Cr and δ^{53}Cr values of main-group aubrites, suggesting the mass-independent data are not influenced by the mass-dependent fractionation. The different δ^{53}Cr values between the three groups of enstatite achondrites suggests they formed from different processes, which is consistent with their different ε^{54}Cr values suggesting at least three enstatite chondrite parent bodies. **Figure 2b**. Mg#-δ^{53}Cr values for aubrites. Except Shallowater and Itqiy, all main-group aubrites have high Mg# (98-100%). The three enstatite achondrite parent bodies have correlated Mg# and δ^{53}Cr values. **Figure 2c.** 1/Cr-δ^{53}Cr values of aubrites. The uncertainty of Cr contents is estimated as 5% (2σ). There is no correlation between 1/Cr and δ^{53}Cr. The reason for the lack of mixing correlation here could be the sample heterogeneity and secondary processes (most likely impact). **Figure 2d.** δ^{56}Fe-δ^{53}Cr values for aubrites. The δ^{56}Fe isotope data are from Wang et al., (2011), and the data with multiple measurements are averaged with 2SD uncertainty. The correlation between δ^{56}Fe and δ^{53}Cr values for main-group aubrites should reflect a mixing between sulfide-metal and silicates. The sulfide-metal are rich in both isotopically light Cr and Fe, while the silicates possess isotopically heavy Cr and Fe.

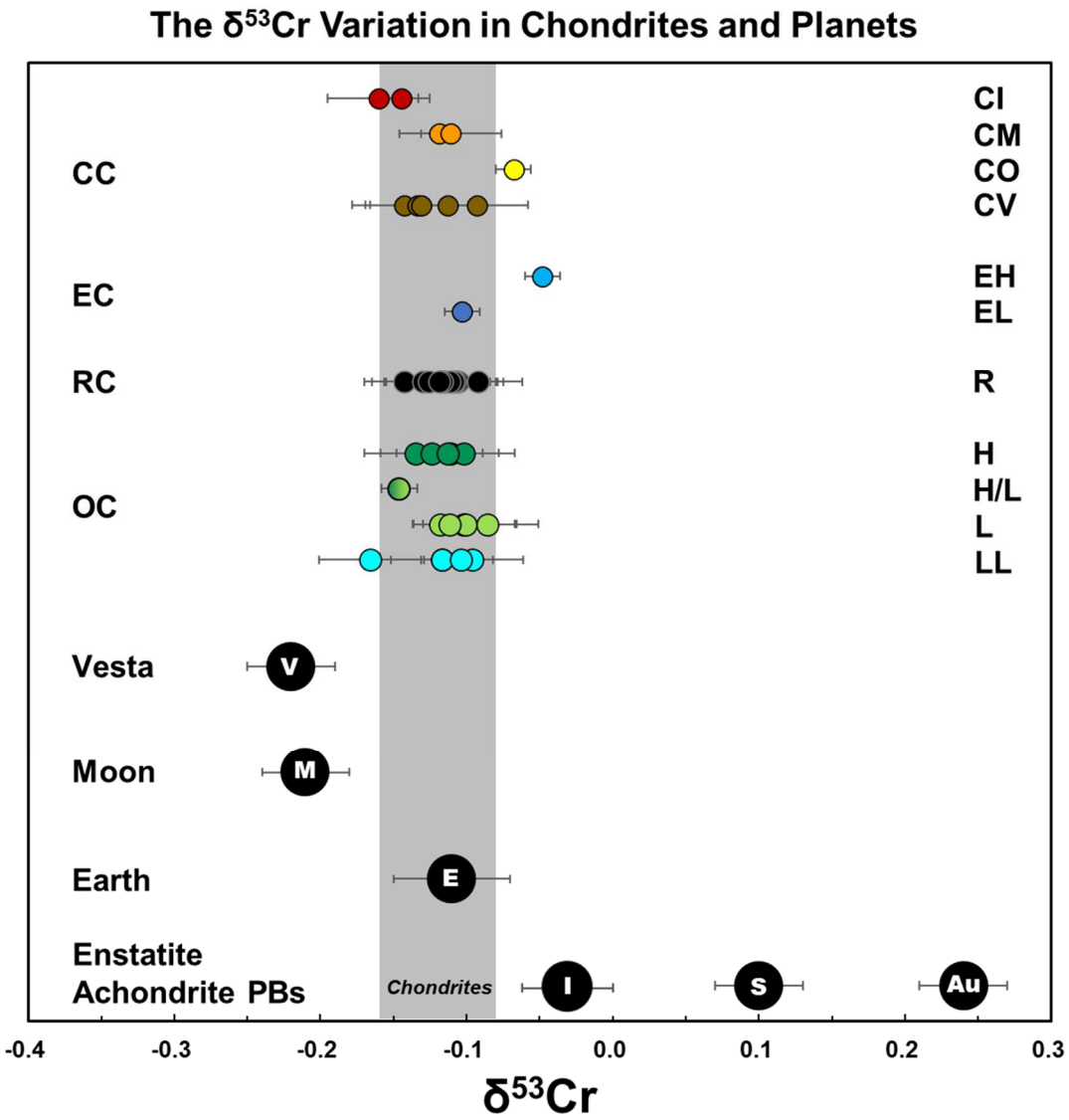


Figure 3. Comparison of the δ^{53}Cr variations amongst chondrites, achondrites and the Earth-Moon system. The small colorful circles are chondrites, while the big black circles represent the Earth, Moon, Vesta and enstatite achondrite parent bodies. The gray bar defines the average δ^{53}Cr values (-0.12 ± 0.04; 2SD, N =42) of all the chondrites, and the three enstatite achondrite parent bodies have distinguished mass-dependent Cr isotope compositions (δ^{53}Cr values): 0.24 ± 0.03 ‰, 0.10 ± 0.03 ‰ and -0.03 ± 0.03 ‰ for main-group aubrite (Au), Shallowater (S) and Itqiy (I) parent bodies respectively, which is consistent their ε^{54}Cr characteristics. The δ^{53}Cr difference between enstatite achondrite parent bodies and chondrites result from a sulfur-rich core formation. Itqiy and Shallowater parent bodies have lower δ^{53}Cr values than main-group aubrites, which may result from more sulfides-metal contents (lower Mg#) in Shallowater and Itqiy. This is consistent with the mixing models in figure 2d. Literature data sources: chondrites (Bonnand et al., 2016b; Schoenberg et al., 2016), Earth (Jerram et al., 2020; Schoenberg et al., 2008; Sossi et al., 2018), Moon (Bonnand et al., 2016a; Sossi et al., 2018) and HEDs-Vesta (Zhu et al., 2019c).

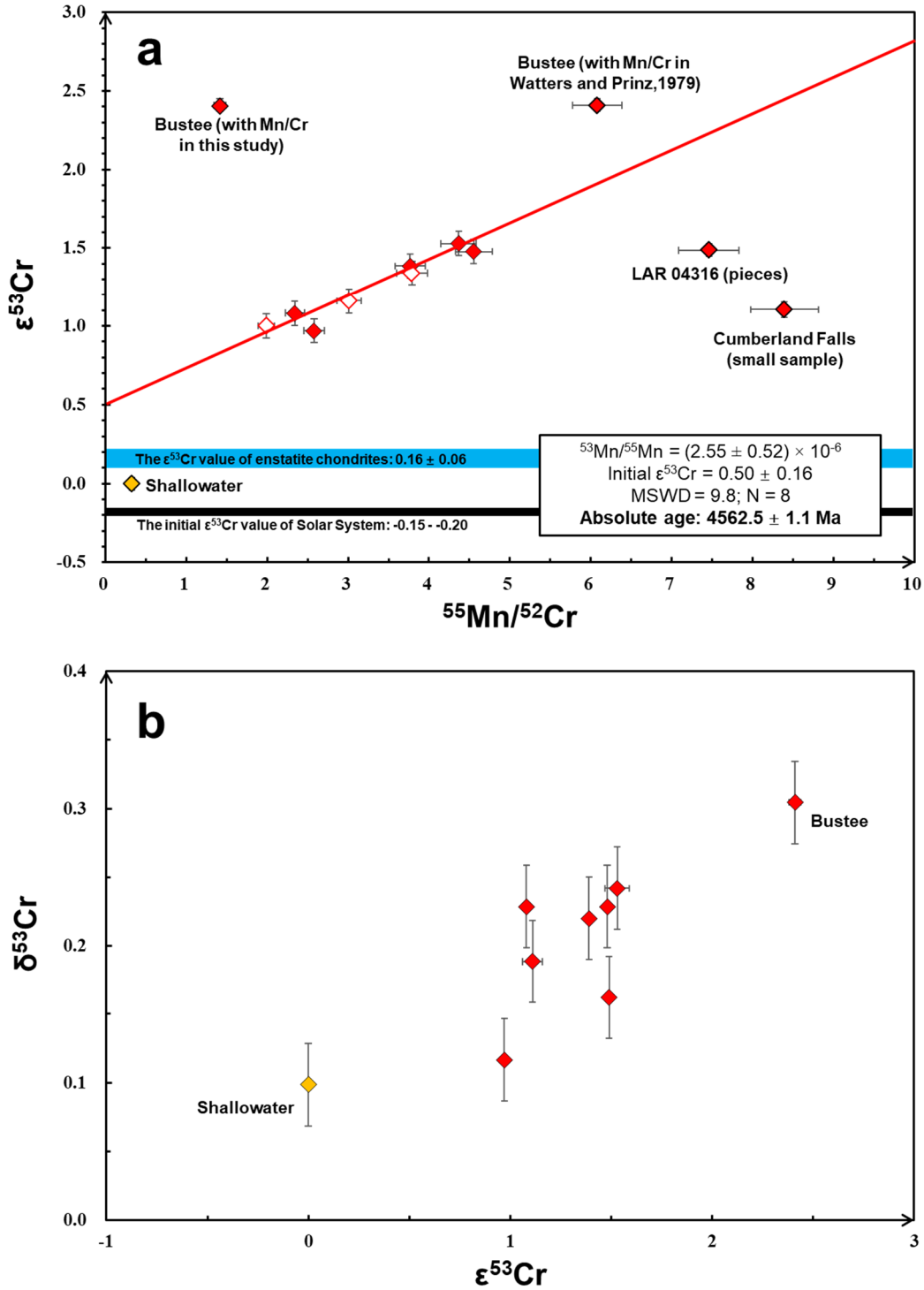


Figure 4a. The ^{53}Mn-^{53}Cr isochron for bulk aubrites, the open diamonds are the Cr isotope data from Shukolyukov and Lugmair (2004). This red correlation line does not include Shallowater, Bustee, LAR 04316 (pieces) and Cumberland Falls (small sample) from this study, due to the different parent bodies and sample heterogeneity. The blue and black bars are the ε^{53}Cr value of enstatite chondrites and the initial ε^{53}Cr value of Solar System that is estimated by D'Orbigny angrite, CI chondrite and chondrule precursors (Göpel et al., 2015; Trinquier et

al., 2008b; Zhu et al., 2019a). The ^{53}Mn-^{53}Cr correlation line is controlled by co-genetic sulfide and silicates, which also holds the time significance reflecting the differentiation of main-group parent body and crystallization of sulfide and silicates. More importantly, the elevated initial ε^{53}Cr value (0.50 ± 0.16, 2σ; higher than that of the Solar System and enstatite chondrites), expressed by the intercept of y axis, could be caused by the early S-rich core formation process. **Figure 4b.** The δ^{53}Cr values correlate with the ε^{53}Cr values that mainly reflect the true Mn/Cr ratios (since there is a large Mn/Cr heterogeneity in aubrites). This correlation indicates that the aubrites reflect mixing of lithologies with more abundant Cr-rich sulfide (low Mn/Cr, low ε^{53}Cr and low δ^{53}Cr) and lithologies with Cr-poor silicates (high Mn/Cr, high ε^{53}Cr and high δ^{53}Cr).

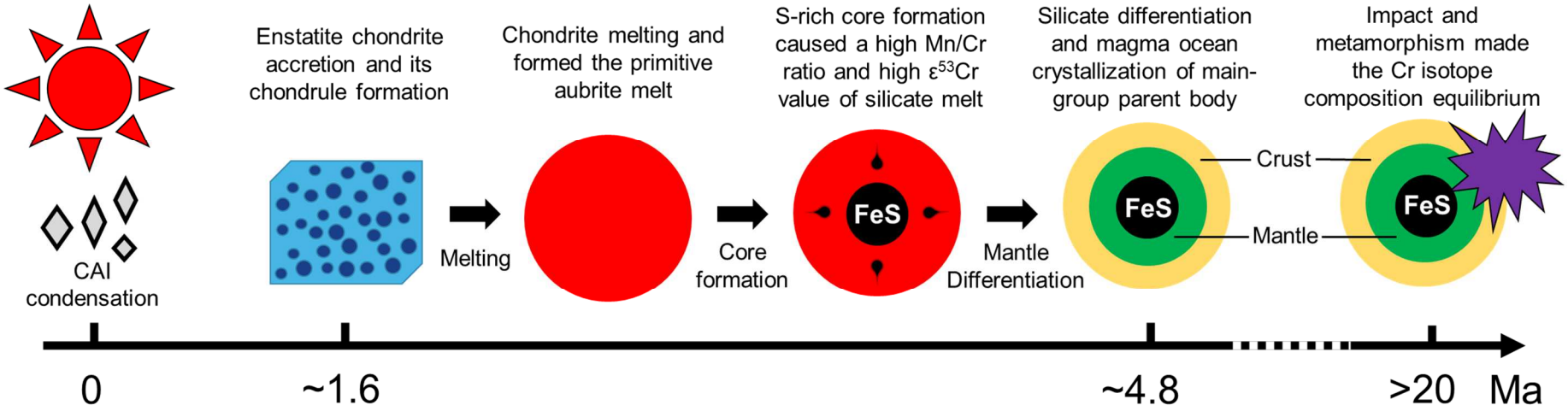


Figure 5 The timeline of evolution of the main-group aubrite parent body. The Ca-Al-rich inclusion condensation marked the formation of the Solar System (t_0, 4567.30 ± 0.16 Ma) (Connelly et al., 2012). Then, enstatite chondrite chondrules formed at 4565.7 ± 0.7 Ma (precursor age), i.e. ~1.6 Ma after t_0 (Zhu et al., 2020a), and the accretion of enstatite chondrites is estimated as ~1.8 Ma after t_0 (Alexander et al., 2018). These two ages can potentially represent the accretion time of enstatite chondrites. After that, enstatite chondrites experienced a melting event and formed the primitive aubrite melt. At this stage, this melt has chondritic Mn/Cr ratio. Then, the sulfur-rich core formation process changed the composition of the primitive melt, making the core rich in Cr and poor in Mn (Berthet et al., 2009), while the silicate melt possessing high Mn/Cr ratio. In this way, after the decay of ^{53}Mn, the silicate melt has a high ε^{53}Cr value that is the source of main-group aubrites. It furthermore experienced a silicate differentiation and mineral crystallization that fractionates the Mn/Cr ratios in main-group aubrites. The ^{53}Mn-^{53}Cr system recorded the age of this event, 4562.5 ± 1.1 Ma (i.e. 4.8 ± 1.1 Ma after CAIs), which indicates the differentiation time of main-group aubrite parent body. Finally, after extinction of ^{53}Mn (around eight times of the half-life of ^{53}Mn) , i.e. ~20 Ma after t_0, there should be a metamorphosed process, mostly caused by impact, redistributed the Mn and Cr in individual aubrites and reset the internal ^{53}Mn-^{53}Cr isochrons.

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