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ORIGINAL ARTICLE

The vanadium isotopic composition of L ordinary chondrites

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Abstract Stable isotopic data of meteorites are critical for understanding the evolution of terrestrial planets. In this study, we report high-precision vanadium (V) isotopic compositions of 11 unequilibrated and equilibrated L chondrites. Our samples show an average $\delta^{51}V$ of − 1.25‰ ± 0.38‰ (2SD, n = 11), which is ~ 0.5‰ lighter than that of the bulk silicate Earth constrained by mantle peridotites. Isotopic fractionation in type 3 ordinary chondrites vary from − 1.76‰ to − 1.29‰, whereas the $\delta^{51}V$ of equilibrated chondrites vary from − 1.37‰ to − 1.08‰. $\delta^{51}V$ of L chondrites do not correlate with thermal metamorphism, shock stage, or weathering degree. Future studies are required to explore the reason for V isotope variation in the solar system.

Keywords V isotopes · L ordinary chondrites · Variation

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1 Introduction

Meteorites are the only easily accessible material with "fossil evidence" of the different evolutionary stages of the solar system and Earth (Hofmann 2010). Stable isotopic data provide an important link between meteorites and the origin of Earth (Schiller et al. 2018; Elardo and Shahar 2017; Dauphas 2017; Burkhardt et al. 2017). From Earth's inception in a proto-planetary disk to accretion and differentiation, multiple processes (e.g. condensation and accretion, evaporation, post-accretional separation) potentially cause stable isotope fractionation (Clayton 2003; Qin et al. 2010; Simon and Depaolo 2010; Warren 2011), resulting in geochemical and isotopic heterogeneity in meteorites. Because primitive chondrites are considered as the most pristine rocks of the solar system without parental body differentiation processes (Huang and Jacobsen 2016; Teng et al. 2010; Valdes et al. 2014; Jacobsen and Wasserburg 1980; Ciesla and Charnley 2006), their stable isotopic data, especially refractory elemental isotopes, can be used to trace the genetic link between meteorites and Earth.

Ordinary chondrites are the most abundant type (> 80% of all witnessed chondritic falls) of primitive chondrites (Graham et al. 1987). In comparison with other types, ordinary chondrites show broader and more systematic changes in composition and structure (Hutchison 2004). Furthermore, the most pristine type 3 ordinary chondrites may retain nebula information, providing clues about the formation of stars and asteroids (Wasson 1972). Thus, a number of stable isotopic systems have been studied in ordinary chondrites.

Vanadium (V) is a refractory element with a half mass condensation temperature of 1427 K (Huang et al. 2015; Lodders 2003). The V isotopic compositions (reported as

$\delta^{51}V = [(^{51}V/^{50}V)_{sample}/(^{51}V/^{50}V)_{AA} - 1] \times 1000‰)$ of early solar system materials may represent initial solar information and shed important light on terrestrial planets' early histories. However, V isotopic data for meteorites are still rare in the literature because of analytical challenges, mainly: (1) with only two stable isotopes (^{51}V and ^{50}V), the double spike strategy is infeasible for V isotopic measurements. Instead, a standard solution-sample bracketing technique needs to be used for precise isotope measurement. This method requires a near 100% yield of V after the total procedure in order to eliminate isotope fractionation during chemical purification. (2) Isobaric interferences (^{50}Cr and ^{50}Ti) on ^{50}V require complete chemical separation of V from those matrix elements. (3) The difference in natural abundance between ^{51}V and ^{50}V is ~ 410 fold, making it difficult to simultaneously measure the two isotopes with sufficient signal and stability. (4) Multi-atom molecular interference (e.g., $^{36}Ar^{14}N^+$, $^{36}Ar^{16}O^+$, $^{38}Ar^{14}N^+$) can also produce erroneous isotope data.

The pioneering work of V isotopes on extraterrestrial materials was conducted in the 1960s and 1970s. However, the uncertainty of V isotopic data in these studies was up to ± 1%—too large to resolve potential variation between meteorites and Earth (Balsiger et al. 1969; Pelly et al. 1970; Lipschutz et al. 1971; Balsiger et al. 1976). With the development of multiple–collector inductively coupled plasma–mass spectrometry (MC–ICP–MS) (Wu et al. 2016; Nielsen et al. 2011, 2016; Prytulak et al. 2011), Nielsen et al. (2014) reported V isotopic data of chondrites and achondrites with a uniform $\delta^{51}V = -1.7‰ \pm 0.2‰$ (2SD). In comparison with bulk silicate Earth (BSE), meteorites were depleted in ^{51}V by about 1‰ (Nielsen et al. 2014). Nielsen et al. (2014) discussed several possible reasons for such considerable fractionation and suggested a early Solar System irradiation origin for the BSE-meteorite V isotopic offset (Nielsen et al. 2014). This hypotheses, however, is difficult to reconcile with the compositional variation of chondrites, which argues against a well-mixed nebula during formation of the solar system (Nielsen et al. 2014). More recent studies exhibited more V isotopic variation than previously indicated (Nielsen et al. 2015, 2017). Sossi et al. (2017) reported 4.4‰ ^{50}V excesses in calcium-aluminum–rich inclusions (CAIs) relative to the bulk chondrite, and co-variation of ^{50}V and ^{10}Be abundance (Sossi et al. 2017). As ^{10}Be was formed in the solar system by irradiation (Gounelle et al. 2008; McKeegan et al. 2000), such a co-variation was interpreted to reflect irradiation processes. If the irradiation in CAI is responsible for the light V isotopes in meteorites, chondrites with different proportions of CAIs should have variable V isotopic compositions. However, V isotope data for chondrites are still rare. Here, we report the V isotopic composition of 11 L chondrites to further constrain V isotopic composition of chondrites. We also explore possible drivers (such as thermal metamorphism, shock stages, and terrestrial weathering) of V isotopic variation.

2 Samples and methods

2.1 Sample descriptions

Based on the narrow ranges in bulk chemical composition (Kallemeyn et al. 1989), oxygen isotopic composition (Clayton et al. 1991), chondrule size (Rubin 2000; Rubin et al. 2008), and oxidation state (McSween and Labotka 1993), ordinary chondrites are subdivided into three distinct groups: H, L, and LL (Hutchison 2004). The 11 L chondrites in this study included two observed falls (Heyetang and Xinglongquan) and nine finds, which were provided by the Polar Research Institute of China. According to their petrographic and mineralogical characteristics, the 11 meteorites were divided into specific petrologic type, shock stage, and weathering degree. Petrologic type, weathering grade, and shock classification were determined through examination of polished thin sections under optical microscope. Petrologic grades of the samples in this investigation are defined according to Van Schmus and Wood (1967), degrees of shock stages according to Stöffler et al. (1991), and weathering degrees according to Wlotzka (1993). Detailed information of each meteorite can be found on the meteorite bulletin site (http://www.lpi.usra.edu/meteor/metbull.php). Their petrographic types, impact degrees, and weathering grades are also shown in Table 1. Among these samples, Heyetang and Xinglongquan are unequilibrated ordinary chondrites (UOCs); the others are equilibrated ordinary chondrites (EOCs).

The Heyetang sample displays typical type 3 rock characteristics with very sharply defined chondrules and clastic opaque matrix (Fig. 1g). In contrast, the poorly defined chondrules in GRV 021475 and GRV 021802 and their matrix with coarse-grained secondary feldspars (Fig. 1b, h) are characteristic of EOCs, and the two meteorites are classified as type 6.

Under optical microscope, thin sections of GRV 052904 displayed normal extinction, and this sample was classified as shock stage S1 (Fig. 1d). GRV 021491 was classified as shock stage S4 with many reticular shock veins and shock melting pockets (Fig. 1e). GRV 052483 was classified as an L-related impact-melt rock with substantive glassy matrix and metal in the form of molten drops and ringwoodites, which indicate that this sample was subjected to strong shock (Fig. 1f).

Table 1 Vanadium isotopic compositions of the meteorite samples

Sample	Petrologic type	Shock stage	Weathering degree	$\delta^{51}V$	2SD	n
GRV 052483	L-imp melt		W1	− 1.08	0.06	3
GRV 052483-R	L-imp melt		W1	− 1.07	0.09	3
GRV 052904	L6	S1	W1	− 1.15	0.04	3
GRV 052904-R	L6	S1	W1	− 1.17	0.05	3
GRV 021491	L6	S4	W1	− 1.37	0.04	3
GRV 051869	L6	S4	W2	− 1.19	0.01	3
GRV 021475	L6	S2	W1	− 1.34	0.04	3
GRV 021802	L6	S2	W1	− 1.18	0.06	3
GRV 021673	L5	S3	W1	− 1.18	0.02	3
GRV 021786	L5	S3	W1	− 1.12	0.06	3
GRV 052076	L4	S2	W3	− 1.13	0.05	3
Xinglongquan	L3	S1	W0	− 1.76	0.05	3
Heyetang	L3	S2	W1	− 1.29	0.05	3
Geostandards						
BIR-1				− 0.92	0.12	3
BCR-2				− 0.76	0.05	3

n number of replicate analyses. GRV 052483-R and GRV 052904-R are the replicated samples of GRV 052483 and GRV 052904, respectively. All data from this study are expressed as ± 2SD (standard deviation)

The effect of weathering on chondrites is texturally characterized by the progressive replacement of pre-existing Fe^0 and Fe^{2+} minerals by Fe-oxides/oxyhydroxides and the appearance of Fe-oxide/oxyhydroxide veins cross-cutting primary minerals (Saunier et al. 2010; Crozaz and Wadhwa 2001). The backscattered electron image (Fig. 1a) shows that more than 70% of the metal in GRV 052076 has been oxidized and limonite is distributed throughout; the sample's weathering grade was classified as W3. On the contrary, metals in GRV 021475 and GRV 021802 are very fresh (Fig. 1c, b), leading to a weathering grade classification of W1.

2.2 Analytical methods

The samples were small fragments free from significant weathering products as sawn from the larger samples. Before cutting, small fragments (approximately 200–300 mg) were inspected to ensure they were not dominated by a large chondrule or metal-sulfide grain, as these may result in isotopic bias (Needham et al. 2009). Then, the chunks of ordinary chondrite were ultrasonically cleaned in distilled water to remove possible external contamination. The samples were then powdered in an agate mortar.

Vanadium isotopic data were measured at the CAS Key Laboratory of Crust-Mantle Materials and Environments at the University of Science and Technology of China (USTC), Hefei. Procedures for sample dissolution, column chemistry, and instrumental analysis are similar to those reported in previous studies (Wu et al. 2016, 2018) and a brief description is given below. Sample powders containing 6–10 μg V were digested in a 3:1 mixture of double-distilled concentrated HF-HNO_3 in Savillex screw-top beakers by heating at 120 °C on a hotplate for 3–4 days. Samples were evaporated to dryness and further dissolved in aqua regia to remove fluorides. We used a HPA-S (Anton Parr®) with 0.5 mL double-distilled concentrated HNO_3 and 1.5 mL double-distilled concentrated HCl to ensure the complete dissolution of all minerals. The samples were dissolved at 320 °C and 100 atm for 16 h. The sample solution was then dissolved in 1 mL 1-mol·L^{-1} HNO_3 for chromatography.

The chemical purification of V was achieved by four steps of cation-exchange column procedure and two steps of anion-exchange column procedure modified from Wu et al. (2016). The first anion-exchange column (Bio-Rad AG1-X8, 200–400 mesh) was used to remove Fe; the second cation-exchange resin column (Bio-Rad AG50 W-X12, 200–400 mesh) was used to remove Mg. The third and fourth cation-exchange resin columns (Bio-Rad AG50 W-X12, 200–400 mesh) were used to remove Ti and some other major matrix elements (e.g., Al, Ca, Mn, and Cr). The last two separation steps with anion-exchange columns (AG1-X8, 200–400 mesh) were to remove residual matrix compounds (such as K, Na, Mg, and Cr). The total yield of chemical procedures was better than 99%. The total procedural blank of V was less than 10 ng, negligible for the amount of V.

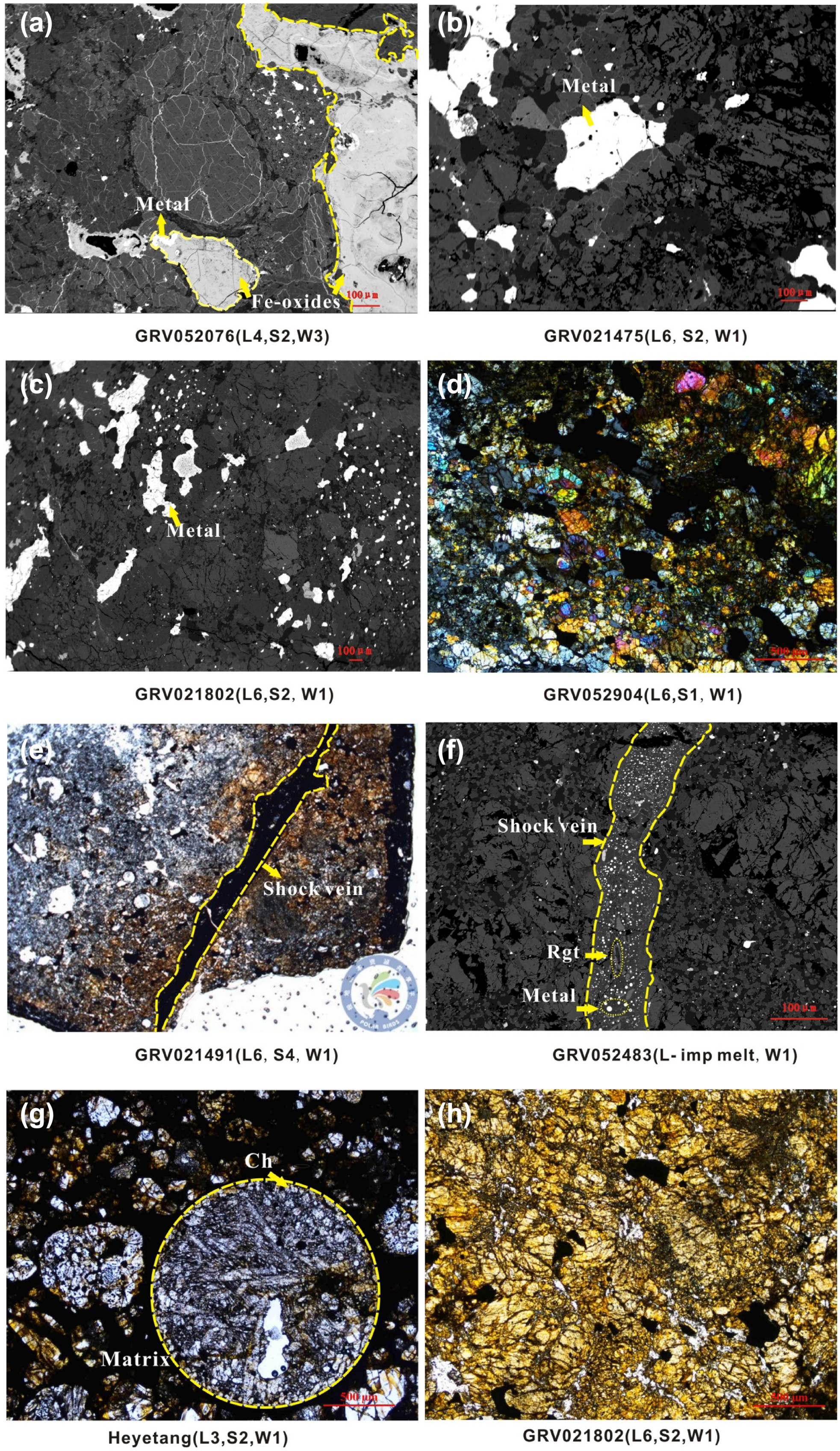

Fig. 1 Optical photomicrograph and backscattered electron images show features of thermal metamorphism, shock stage, and weathering stage of our meteorite samples. **a** Most of the Fe–Ni metal in GRV 052076 has altered to Fe-oxides, suggesting a strong degree of weathering. The Fe-Ni metal in **b** and **c** are not altered. **d** The olivines in GRV 052904 display normal extinction, showing no traces of impact. **e** GRV 021491 shows shock features with shock veins, and this picture is from the Resource-sharing Platform of Polar Samples. **f** GRV 052483 shows highly impacted features with shock glass and newly formed high-pressure minerals (ringwoodite). **g** Heyetang shows well-preserved chondrules and clastic opaque matrix. **h** The contours of the chondrules in GRV 021802 are unclear and the matrix is crystallized and coarse granular. Rgt: ringwoodite; Ch: chondrule

Vanadium isotopic ratios were measured on Neptune Plus MC-ICP-MS in medium-resolution mode (m/ΔM > 5500) with Jet sampling and Ni X-skimmer cones. ^{49}Ti, ^{50}V, ^{51}V, ^{52}Cr, ^{53}Cr, ^{54}Fe, and ^{56}Fe were measured in static mode on L4, L2, L1, C, H1, H3, and H4 Faraday cups, respectively. Vanadium isotope ratios were measured on the left shoulder to avoid interferences from Ti isotopes (^{48}Ti and ^{49}Ti), Cr isotopes (^{52}Cr and ^{53}Cr), and other isobaric effects. Standard bracketing with V standard (USTC-V) was used to correct instrumental mass bias. The $\delta^{51}V$ value of USTC-V relative to the Alfa Aesar (AA) standard solution measured was 0.07‰ ± 0.07‰ (2SD, n = 112) (Wu et al. 2016). Isotopic data are reported using the delta (δ) notation relative to AA standard solution:

$$\delta^{51}V = \left[\left({}^{51}V/{}^{50}V\right)_{sample} / \left({}^{51}V/{}^{50}V\right)_{AA} - 1\right] \times 1000‰$$

Accuracy and precision of isotopic measurements were assessed by analyzing reference materials, including BCR-2 and BIR-1. The long-term external precision of $\delta^{51}V$ was better than 0.08‰ (2SD). The measured standard values (Table 1) agreed well with published data within error (Wu et al. 2016). $\delta^{51}V$ of replicates were also consistent within error, demonstrating the reliability of our data. Notably, the data quality in this study is better than that in previous studies on meteorites.

3 Results

Vanadium isotopic data are reported in Table 1 and plotted in Figs. 2 and 3. The $\delta^{51}V$ of the L chondrites range from − 1.76‰ ± 0.05‰ to − 1.08‰ ± 0.06‰, with an average of − 1.25‰ ± 0.38‰ (2SD, n = 11). With the exception of Xinglongquan (the lightest sample, with $\delta^{51}V$ = − 1.76‰ ± 0.05‰), the majority of our samples are in the range of − 1.05‰ to − 1.35‰ (the shaded area in Fig. 2). The $\delta^{51}V$ data of the two UOC samples vary from − 1.76‰ to − 1.29‰ whereas EOCs vary from − 1.37‰ to − 1.08‰.

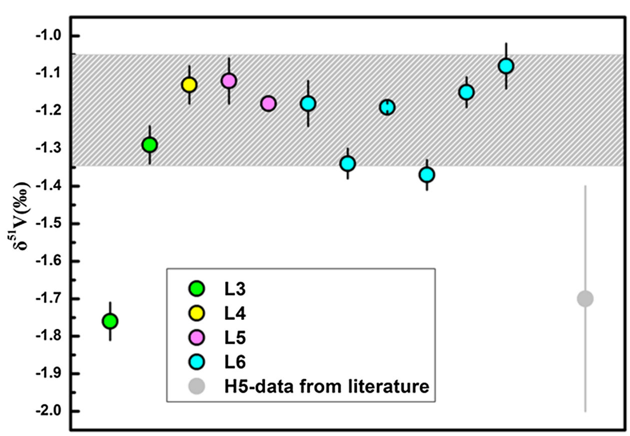

Fig. 2 Vanadium isotopic compositions of L chondrites. H5 data from Nielsen et al. (2014)

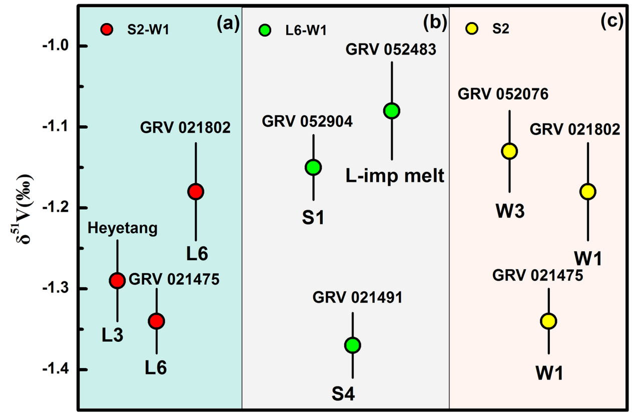

Fig. 3 $\delta^{51}V$ variation with **a** thermal metamorphism, **b** shock stage, and **c** weathering stage. **a** The three representative samples have the same shock stage (S2) and weathering degree (W1), but different thermal metamorphism; **b** Three samples have the same thermal metamorphism (L6) and weathering degree (W1), but different shock stages; **c** The representative samples have the same shock stage (S2) and different weathering degrees regardless of their thermal metamorphism

4 Discussion

Nielsen et al. (2014) reported the first V isotope data for ordinary chondrite, Plainview (H5), as $\delta^{51}V$ = − 1.7‰ ± 0.3‰. Later, an LL chondrite, Chelyabinsk, was reported to have an intermediate V isotope value between estimated BSE (− 0.7‰ ± 0.2‰) and previously published homogeneous meteorite data (− 1.7‰ ± 0.2‰) (Nielsen 2015). The latest 49th Lunar and Planetary Science Conference proceedings reported that chondrites preserve a limited range of $\delta^{51}V$ from − 1.05‰ to − 1.35‰ (Nielsen 2018). Our data have a more extensive range of $\delta^{51}V$ from − 1.08‰ to − 1.76‰, confirming that the meteorites do not represent a homogeneous isotopic composition (Nielsen et al. 2015). Our results are consistent with prior studies in that meteorites measured thus far exhibit much lighter isotopic composition than the best estimate for BSE. We found that V isotope composition of BSE is heavier than that of the average L chondrites by about 0.5‰, much smaller than the 1‰ reported previously (Nielsen et al. 2014).

4.1 Effects of thermal metamorphism, shock stage, and terrestrial weathering

Petrologic and geochemical evidence show that most ordinary chondrites endured thermal metamorphism on chondrites' parent bodies (Kessel et al. 2007). GRV 021475, Heyetang, and GRV 021802 share the same impact grade and weathering degree (Table 1). Heyetang has typical type 3 rock characteristics while GRV 021475 and GRV 021802 generally show type 6 characteristics. As shown in Fig. 3, $\delta^{51}V$ is not simply correlated with thermal metamorphism. There is no correlation between $\delta^{51}V$ and petrologic type of the meteorites in this study, indicating that thermal metamorphism in the parent bodies may not affect V isotopes. This can likely be explained by V's refractory nature with a half-mass condensation temperature (T_c) of 1427 K, much higher than the commonly assumed peak range of metamorphic temperatures (600–950 °C). The isotopic composition of elements with T_c close to metamorphic temperatures, such as Zn (T_C = 726 K) and Cd (Tc = 652 K), may be associated with thermal metamorphism (Luck et al. 2005; Schmitt 2016), but this is not the case for V isotopes.

There is no clear relationship between shock stage and $\delta^{51}V$ either (Fig. 3). Petrologic studies classified the shock stage of GRV 052904 as S1, and GRV 021491 as S4. GRV 052483 is an L-related impact-melt rock. GRV 052483 and GRV 021491 show distinct $\delta^{51}V$ values of − 1.08‰ ± 0.06‰ and − 1.37‰ ± 0.04‰, respectively, whereas GRV 052483 and GRV 052904 have the same $\delta^{51}V$ within error. Therefore, we suggest that V isotopic fractionation is not associated with the heating and volatilization caused by impact events.

In previous studies, a preferential loss of the heavy isotope in meteorites has been posited to occur during terrestrial weathering (Luck et al. 2005; Paniello et al. 2012). However, in this study we observe no systematic variation of $\delta^{51}V$ with different weathering stages between the falls and the finds (Fig. 2). Furthermore, V isotopic composition of GRV 052076, GRV 021475, and GRV 021802 clearly show no correlation with weathering stage (Fig. 3). This is in consistent with the observation of terrestrial rocks that aqueous weathering has limited effects on V isotopes (Prytulak et al. 2013).

4.2 Reasons for diversity of vanadium isotopic composition of meteorites

V isotopes have long been thought a good tracer of early solar system irradiation that produced ^{50}V from target nuclei of Ti and Cr (Balsiger et al. 1969; Pelly et al. 1970). Despite recent progress showing that early solar system irradiation could have caused enrichment of light V isotope (^{50}V) among CAIs, other recent advances have shown the existence of CAIs in carbonaceous chondrites has little impact on bulk V isotope composition (Nielsen et al. 2018). Moreover, CAIs in unequilibrated ordinary meteorites (< 0.1 vol.%) are rarer and smaller than those in carbonaceous chondrite (up to 15 vol.%) (Hutchison 2004). Thus, the lightest $\delta^{51}V$, which is found in Xinglongquan, cannot be due to the existence of CAIs.

Because of the existence of only two V isotopes in nature, it is difficult to relate V isotope variations directly to a singular process. A strong positive correlation between V isotopes and ^{54}Cr nucleosynthetic isotope anomalies have been reported in carbonaceous chondrites, which implies a common origin for the variation of the two isotope systems (Nielsen et al. 2018). Beacause $\delta^{51}V$ in our samples show a variation while ordinary chondrites exhibit a common $^{54}Cr/^{52}Cr$ deficit of 0.4ε (Qin et al. 2010), we can not directly relate vanadium isotopic variation to nucleosynthetic isotope anomalies. The limited vanadium isotope database for meteorites presented are far from sufficient to explain the isotopic variations. Only further and more detailed V isotope investigations of extraterrestrial material will enable a more unambiguous evaluation of the origin of V isotope variation in meteorites and on Earth.

5 Conclusions

This study reports V isotope composition for 11 L chondrites. $\delta^{51}V$ of UOCs (type 3 rock) vary from − 1.76‰ to − 1.29‰, while EOCs cluster tightly with $\delta^{51}V$ ranging from − 1.37‰ to − 1.08‰. As opposed to previous research that meteorites are enriched in ^{50}V compared with BSE by 1‰, our L chondrite samples are enriched in ^{50}V by about 0.5‰ relative to BSE. Parent body metamorphism, shock stage, and terrestrial weathering do not show clear relationships with $\delta^{51}V$, implying little impact on V isotope fractionation. More study is required to distinguish the effects of high-energy irradiation and nucleosynthetic heterogeneity on V isotopic variation.

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