



# Vanadium isotopic difference between the silicate Earth and meteorites



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## ABSTRACT

It has been argued that the stable isotopic composition of the element vanadium (V) provides a potential indicator of the effects high-energy irradiation early in Solar System development. Such irradiation would produce enrichment in the minor isotope,  $^{50}\text{V}$  compared with the 400 times more abundant  $^{51}\text{V}$  (Gounelle et al., 2001; Lee et al., 1998). Here we show that the vanadium isotopic composition of the silicate Earth is enriched in  $^{51}\text{V}$  by  $\sim 0.8\%$  compared with carbonaceous and ordinary chondrites as well as achondrites from Mars and the asteroid 4 Vesta. Although V is depleted by core formation, experiments reveal no isotopic fractionation between metal and silicate that could account for the observed difference in V isotope composition between terrestrial and extraterrestrial materials. Nucleosynthetic provenance of the terrestrial vanadium isotope offset is inconsistent with anomalies of other nucleosynthetically produced isotopes in bulk meteorites, which are more variable than vanadium (Burkhardt et al., 2011; Carlson et al., 2007; Trinquier et al., 2009). Furthermore, V isotopes are unlikely to have been affected by volatilization, parent body alteration or impact erosion of Earth's surface. Therefore, the cause of the isotopic difference is unclear. One possibility is that Earth's isotopically heavier V reflects a deficit in material irradiated during the initial stages of Solar System formation. Whatever the cause, the terrestrial deficit in  $^{50}\text{V}$  implies that bulk Earth cannot be entirely reconstructed by mixtures of different meteorites.

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## 1. Introduction

It is believed that the earliest phases of the Sun's evolution were characterized by frequent violent bursts of high-energy particles (Shu et al., 1997). However, the consequences for the synchronous formation and distribution of the first solids in the Solar System are the topic of much debate. In a series of papers Shu and co-workers developed a physical model in which calcium aluminium rich inclusions (CAIs) and possibly chondrules were formed close to the Sun in a high-energy irradiation environment and subsequently transported to the Asteroid Belt via the so-called X-wind (Lee et al., 1998; Shu et al., 1996, 1997, 2001). The X-wind is a specific type of magnetocentrifugal outflow of material, which is a common feature of young stellar objects (Coffey et al., 2007). It was also proposed that a host of short-lived radioactive isotopes, such as  $^{10}\text{Be}$ ,  $^{26}\text{Al}$  and  $^{41}\text{Ca}$ , would have formed in this environment, which was suggested could explain the observed abundances of these isotopes in CAIs (Gounelle et al., 2001;

Lee et al., 1998). However, several lines of evidence indicate that the specific physical mechanisms invoked for formation and transport of CAIs, chondrules and short-lived radioactive nuclides (SLRs) are not realistic in an X-wind scenario (Desch et al., 2010; Wood, 2004).

Still, there is compelling evidence to indicate that irradiation was an integral part of the early stages of Solar System evolution both based on astronomical and meteoritic observations (Burnett et al., 1965; Coffey et al., 2007; Feigelson, 2010; Fowler et al., 1962; Jacobsen et al., 2011; McKeegan et al., 2000). Therefore, more information is required before we can obtain a better understanding of the processes that shaped the composition of our Solar System.

The aforementioned calculations of the isotopic consequences of the X-wind model also considered the stable isotope system of vanadium (Gounelle et al., 2001; Lee et al., 1998). Even though the X-wind model is difficult to reconcile with our knowledge of CAIs, chondrules and some SLRs, the effects of Solar System irradiation is still relevant for predicting potential isotopic anomalies in irradiated material. It was shown that significant enrichments of several permil in  $^{50}\text{V}$  would be expected for material processed in an environment with high-energy irradiation (Gounelle et al., 2001; Lee et al., 1998). Therefore, high-precision measurements of V

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isotopes in extraterrestrial material may reveal new information about the irradiation history of the Solar System.

Here we present the first measurements of the vanadium isotopic composition of a number of chondrites and achondrites from Mars and the HED parent body. Our results reveal a striking vanadium isotopic offset between the Earth and extraterrestrial material, which is most consistent with Earth accreting from material that was less irradiated than Mars and the Asteroid Belt. This finding supports the hypothesis that Earth is not made entirely from material with chondritic relative abundances of refractory elements.

## 2. Vanadium isotope background

The element vanadium (V) has two isotopes with masses 50 and 51 and abundances of 0.24% and 99.76%, respectively. The isotope composition of vanadium has long attracted interest from cosmochemists because it was realized that high-energy irradiation has significant potential to produce  $^{50}\text{V}$  mainly from target nuclei of Ti and Cr (Burnett et al., 1965). It was therefore believed that V isotope compositions of meteorites and lunar samples could elucidate the irradiation history of the early solar system and potentially show if irradiated material was heterogeneously distributed within the solar system. However, despite significant analytical endeavors no researchers were able to identify any V isotopic variation outside an analytical uncertainty of about  $\pm 1\%$  (Balsiger et al., 1969, 1976; Pelly et al., 1970).

Recently the first method that produces highly precise and accurate V isotope compositions to an external two standard deviations precision of  $\pm 0.015\%$  was developed (Nielsen et al., 2011; Prytulak et al., 2011). This advance in analytical precision is almost two orders of magnitude superior to earlier efforts and allows the first high-precision investigation of vanadium isotopic variation in meteorites and on Earth. Vanadium isotope compositions are reported as

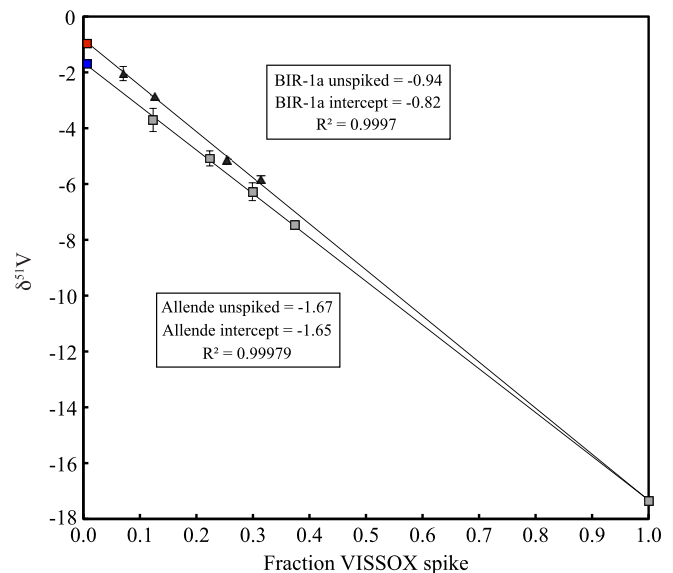
$$\delta^{51}\text{V} = 1000 \times \left[ \left( \frac{^{51}\text{V}}{^{50}\text{V}} \right)_{\text{sample}} - \left( \frac{^{51}\text{V}}{^{50}\text{V}} \right)_{\text{AA}} \right] / \left( \frac{^{51}\text{V}}{^{50}\text{V}} \right)_{\text{AA}}$$

where AA is a V solution standard purchased from Alfa Aesar (Lot #91-092043G) which is defined as  $\delta^{51}\text{V} = 0$ . Thus negative values denote enrichments in  $^{50}\text{V}$ .

## 3. Methods

### 3.1. Vanadium isotopic measurements

Vanadium isotope data were obtained using previously described chemical separation and mass spectrometry methods (Nielsen et al., 2011; Prytulak et al., 2011). About 3–5  $\mu\text{g}$  of V was consumed in each individual isotopic measurement and therefore around 50–100 mg of sample was weighed out for each sample split in order to have enough V for at least one measurement. Samples were digested in Teflon vials in mixtures of HF and  $\text{HNO}_3$  either on a hotplate (achondrites) or in an Anton Parr microwave digestion system (chondrites). These procedures ensured complete digestion of all samples and no residues were encountered for any of the samples investigated, except for the ureilite where minor amounts of graphite were still present after microwave digestion. These residues were removed prior to V separation via centrifugation as it was assumed that graphite contains negligible amounts of V. Briefly, chemical separation is performed with liquid anion exchange chromatography where each sample is subjected to at least five column passes. The procedure is especially designed to remove the elements Cr and Ti quantitatively, because  $^{50}\text{Cr}$  and  $^{50}\text{Ti}$  interfere directly on  $^{50}\text{V}$  during mass spectrometry and are difficult to correct for when either  $^{53}\text{Cr}/^{51}\text{V}$  or  $^{49}\text{Ti}/^{51}\text{V} > 0.00001$



**Fig. 1.** Standard addition tests for Allende and USGS Icelandic basalt BIR-1a in which small known amounts of isotopically fractionated standard VISSOX was added to splits of Allende and BIR-1a (Prytulak et al., 2011). Regression lines with pure VISSOX and VISSOX-sample mixtures yield intercepts that are identical to unspiked samples. This coherence shows that no detectable matrix effects are present when measuring chondrites or basalts, which encompass all sample compositions investigated in this study. Error bars are two standard deviations on repeat measurements of the same solution. Symbols without error bars have uncertainties smaller than the symbol size.

(Nielsen et al., 2011). All samples run during the course of this study exhibited  $^{53}\text{Cr}/^{51}\text{V}$  or  $^{49}\text{Ti}/^{51}\text{V} < 0.00001$ , most with values being below 0.000003. The procedure returns quantitative yields for V and thus eliminates any potential V isotope fractionation during chemical separation. Chemical yield of V during column chemistry was monitored throughout this study by comparing V concentrations (measured on minor splits by ICP-MS) with the amount of V recovered from the column chemistry procedure. All samples were found to have yields of  $100\% \pm 15\%$ . Isotope ratios were measured on a Nu Instruments MC-ICPMS relative to an Alfa Aesar specpure standard (Nielsen et al., 2011).

Several tests were performed in order to assess precision and accuracy of V isotopic measurements on both silicate rocks and chondrites. Specifically, we conducted standard addition tests on Allende and USGS Iceland basalt BIR-1a in which known amounts of an isotopically fractionated standard (VISSOX: Vanadium Isotope Standard Solution OXFord) was added in different proportions to splits of the samples. These standard-sample mixtures were then processed as regular samples and isotope compositions were measured. Fig. 1 shows the mixing lines produced by the standard addition data and how the intercepts of these mixing lines compare with the unspiked samples. It is evident that unspiked samples return values that are identical within error of the mixing line intercepts, which shows that, within the uncertainties of our measurements, there are no systematic matrix effects or inaccuracies that affected the isotope compositions of the meteorites (or terrestrial samples published elsewhere by Prytulak et al., 2013).

Precision was determined via multiple measurements of the same sample where separate splits of the powdered sample were digested and processed through chemistry and then measured individually on the mass spectrometer. In order to detect inconsistencies, all samples in this study were analyzed at least in duplicate and the reproducibility is similar to the long-term precision of USGS reference rock samples and Allende of about 0.1–0.2‰ (Table 1) (Prytulak et al., 2011, 2013).

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