

Nuclear field shift effect as a possible cause of Te isotopic anomalies in the early solar system—An alternative explanation of Fehr et al. (2006 and 2009)

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(Received 22 April 2009; revision accepted 01 September 2009)

Abstract—We explore the possibility that Te isotopic anomalies measured in Ca-Al-rich inclusions (Fehr et al. 2009) and in leachates of carbonaceous chondrites (Fehr et al. 2006) may be due to mass-independent effects controlled by nuclear field shift rather than to nucleosynthetic processes. Fehr et al.'s spectrum of mass-independent anomalies of Te isotopes shows a smooth correlation with mass number and nuclear charge distribution. Ratios of even to odd isotopes, as the $^{125}\text{Te}/^{126}\text{Te}$ ratio used by these authors for normalization are particularly prone to nuclear field shift effects. We show that the alternative normalization of isotopic ratios to $^{130}\text{Te}/^{126}\text{Te}$ strongly reduces the trend of isotopic fractionation with mass number, leaving only ^{125}Te as truly anomalous.

For both normalizations ($^{125}\text{Te}/^{126}\text{Te}$ and $^{130}\text{Te}/^{126}\text{Te}$), Fehr et al.'s results fit the theory of Bigeleisen (1996), which suggests that the nuclear field shift effect can potentially account for the observed Te isotope abundances, as an alternative to nucleosynthetic processes.

We propose that these mass-independent effects may be acquired during accretion of sulfides from the solar nebula.

INTRODUCTION

Fehr et al. (2005) demonstrated the overall lack of mass-independent isotope fractionation of tellurium (Te) in bulk carbonaceous chondrites, ordinary chondrites, and in iron meteorites. In contrast, Fehr et al. (2009) and Fehr et al. (2006) report non mass-dependent effects on Te isotopes. Fehr et al. (2009) found large isotopic anomalies in two coarse-grained calcium-aluminum-rich inclusions (CAIs). CAIs USNM 3529 and USNM 5171 displayed small negative anomalies for $\epsilon^{122}\text{Te}$ (-17 ± 11 and -48 ± 26) and small positive anomalies for $\epsilon^{128}\text{Te}$ (7.1 ± 4.0 and 22 ± 14) and $\epsilon^{130}\text{Te}$ (15.3 ± 7.6 and 43 ± 26) when normalized to $^{125}\text{Te}/^{126}\text{Te}$. In addition, CAIs USNM 3529 displays small negative anomaly for $\epsilon^{124}\text{Te}$ (-9.0 ± 6.3). The authors ascribed the anomalies to nucleosynthetic effects, uncontrolled analytical artifacts or mass-independent isotopic fractionation by field shift effect. Fehr et al. (2006) found smaller Te isotopic anomalies in the leachable components of Orgueil, Murchison and Allende. The authors observed that the “nitric acid leachate of Murchison displays an anomalous $\epsilon^{130}\text{Te}$ of $+3.5 \pm 2.5$ ” and ascribed these anomalies to small excesses of an

r-process component or to small deficits of an *s*-process component at mass 130.

The conventional interpretation (Bigeleisen and Mayer 1947; Urey 1947) of natural variations in the isotopic abundances of oxygen, sulfur, and other light elements regards the nucleus as a point charge and the resulting isotope fractionation varies smoothly with the masses of the isotopes. We contend elsewhere (Fujii et al. 2006a, 2006b) that many isotopic anomalies measured in meteorites can actually be explained by the finite volume of the nuclear charge with, in particular, “staggering” being observed between odd and even atomic masses. Natural isotopic variations of Hg and Tl have also been ascribed to nuclear field shift effects (Schauble et al. 2007). Nuclear field shift creates just another form of thermodynamic isotope fractionation, but the linear dependence of isotopic effects with mass, which is the hallmark of stable isotope geochemistry, is lost.

With the preconception that normalization using two isotopes (^{126}Te and ^{125}Te) with different parity unfortunately propagates the effect of nuclear field shift into the entire spectrum of isotopes, we here demonstrate that the isotopic variation of Te found in CAIs (Fehr et al. 2009) and

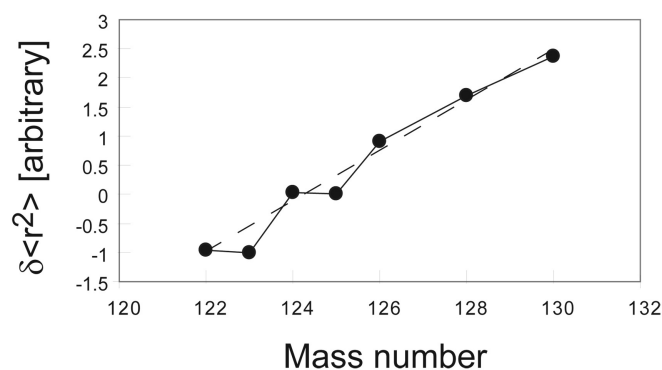


Fig. 1. Changes in nuclear radii for Te isotopes. These changes can be represented as the sum of a linear term (which should not be mistaken for the point charge effect of the standard theory), higher order terms (here negligible), and of odd-even “staggering,” $\delta\langle r^2 \rangle$ (the isotopic difference in the mean-square charge radius), values were calculated from literature root-mean-square charge radii in fm (10^{-15} m) (Aufmuth et al. 1987).

carbonaceous chondrites (Fehr et al. 2006) can be accounted for by Bigeleisen’s (1996) theory of nuclear field shift.

THEORETICAL BACKGROUND ON THE NUCLEAR FIELD SHIFT EFFECT

If nuclei could be considered as electric point charges, the curve describing the intermolecular potential between each isotope of a same element and a particular molecule or ligand would be unique. Zero-point energy would control mass fractionation between species and mass fractionation would, to a first order, vary linearly with mass number. This is the substance of the standard theory of stable isotope fractionation formulated by Urey (1947) and Bigeleisen and Mayer (1947). In contrast, when the charges in the nucleus are distributed over a finite volume, changes in the nucleus configuration, in particular the spatial distribution of nuclear charges, affect the intermolecular potentials, in particular the minimum of vibrational potential curves, from one isotope to another, and therefore give rise to mass-independent isotope fractionation. Bigeleisen (1996) provided the basic equations describing this effect, which in the standard epsilon notation can be recast as:

$$\varepsilon = \left(\frac{hc}{kT} \right) \times \delta\langle r^2 \rangle \times A + \frac{1}{24} \left(\frac{h}{2\pi kT} \right)^2 \times \left(\frac{1}{m} - \frac{1}{m'} \right) \times B \quad (1)$$

where m' and m indicate the masses of the light and heavy isotopes, respectively. The first term on the right-hand side represents the nuclear field shift and most importantly includes the isotopic difference in the mean-square charge radius $\delta\langle r^2 \rangle$ (King 1984). T is the temperature, k and h the Boltzman and Planck constants, respectively, c the velocity of light, and A and B are adjustable constants describing the nuclear field shift effect and the conventional mass effect,

respectively (Bigeleisen and Mayer 1947). Figure 1 shows the changes in mean-square charge radius $\langle r^2 \rangle$ with mass number for Te.

For many elements (Ti, Cr, Zn, Ba, Sm, Nd, Gd, Mo, Te, Sn, and Ru), the existence of mass-independent isotope effects is consistent with this theory. These have been confirmed experimentally by chemical exchange methods (solvent extraction or liquid chromatography) with macrocyclic compounds (Fujii et al. 1998a, 1998b, 1999, 2000, 2001, 2002a, 2002b, 2006b, 2009; Moynier et al. 2008, 2009).

Equation 1 can first be fitted to isotopic data for parameters A and B . When no nuclear field shift effect is considered, it is standard practice to explore potential radiogenic and nucleosynthetic anomalies after normalization to a reference ratio. The advantage of isotopic normalization is that a representation of the anomalies with optimum precision is easily obtained. In particular, Fehr et al. (2006) normalized their Te isotope data to the reference $^{125}\text{Te}/^{126}\text{Te}$ ratio and Fehr et al. (2009) normalized their Te isotope data to both the $^{125}\text{Te}/^{126}\text{Te}$ ratio and the $^{122}\text{Te}/^{124}\text{Te}$ ratio. Since we argue that effects beyond the nuclear point charge should also be considered, we will therefore explore the effect of a particular choice of this reference ratio on the breakdown of isotope fractionation.

DISCUSSION

Figure 2 shows the new data reported by Fehr et al. (2009) for two CAIs, USNM 3529 and USNM 5171 normalized to $^{125}\text{Te}/^{126}\text{Te}$ (Fig. 2a) and to $^{122}\text{Te}/^{124}\text{Te}$ (Fig. 2b). We can check that the normalization to an odd/even couple, $^{125}\text{Te}/^{126}\text{Te}$, leaves a residual correlation with mass number much larger than normalization to two even isotopes, $^{122}\text{Te}/^{124}\text{Te}$.

When isotopic data are normalized to a reference ratio, Fujii et al. (2006a) showed that Equation 1 can be simplified to:

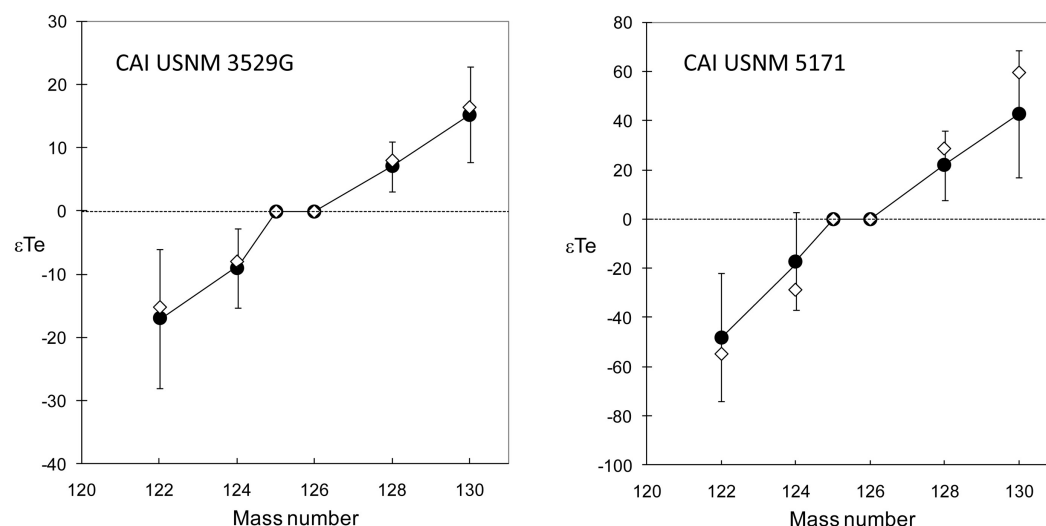
$$\varepsilon = \left[\delta\langle r^2 \rangle_{m_1, m_i} - \frac{m_2(m_i - m_1)}{m_i(m_2 - m_1)} \delta\langle r^2 \rangle_{m_1, m_2} \right] \times a \quad (2)$$

where m_1 and m_2 are the masses of the isotopes used for normalization (e.g., $m_1 = 126$ and $m_2 = 130$); m_i is the mass of the isotope used as nominator (e.g., $m_i = 122, 124, 125, 128$, or 130) and a is an adjustable parameter. Therefore, only one free parameter (a) needs to be adjusted to fit the isotopic data. As shown in Fig. 1, the mean-square charge radius $\langle r^2 \rangle$ of an odd atomic mass number isotope (with an odd number neutrons) is smaller than the value expected from adjacent even atomic mass number isotopes (with an even number neutrons).

By adjusting the parameter a from Equation 2, the isotopic variations of Te can be fitted by the nuclear field-shift

a.

- Fehr et al. (2009) normalized to $^{125}\text{Te}/^{126}\text{Te}$
- ◇ Calculated from the field shift effect theory



b.

- Fehr et al. (2009) normalized to $^{122}\text{Te}/^{124}\text{Te}$
- ◇ Calculated from the field shift effect theory

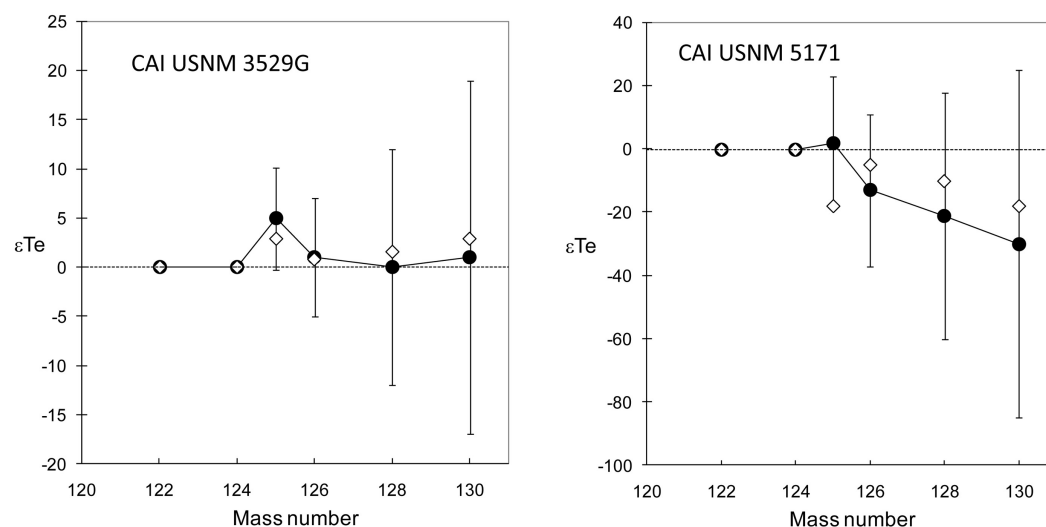


Fig. 2. Isotopic variations of Te in CAIs from Allende (Fehr et al. 2009). a) Normalized to $^{125}\text{Te}/^{126}\text{Te}$ and b) normalized to $^{122}\text{Te}/^{124}\text{Te}$. The calculated symbols correspond to the fit of the measured data by the nuclear field shift theory using Equation 2. ^{120}Te is not shown due to its large analytical uncertainty.

theory for the two normalizations used by Fehr et al. (2009). It should be noted that both the field shift effect and nucleosynthesis models have only one free parameter.

Figure 3 shows the data reported by Fehr et al. (2006) for the leachates of three carbonaceous chondrites, Allende (CV3), Murchison (CM2), and Orgueil (CI) normalized to $^{125}\text{Te}/^{126}\text{Te} = 0.374902$ (Lee and Halliday 1995) as a function

of mass number. As emphasized by Fehr et al. (2006), most normalized values show anomalies that may not be significant with respect to the error bars. The most significant anomaly at mass 130 was explained by these authors as a potential nucleosynthetic effect. The smooth change of the deviation from the reference values across the mass range suggests a different interpretation. It is unlikely that these effects are

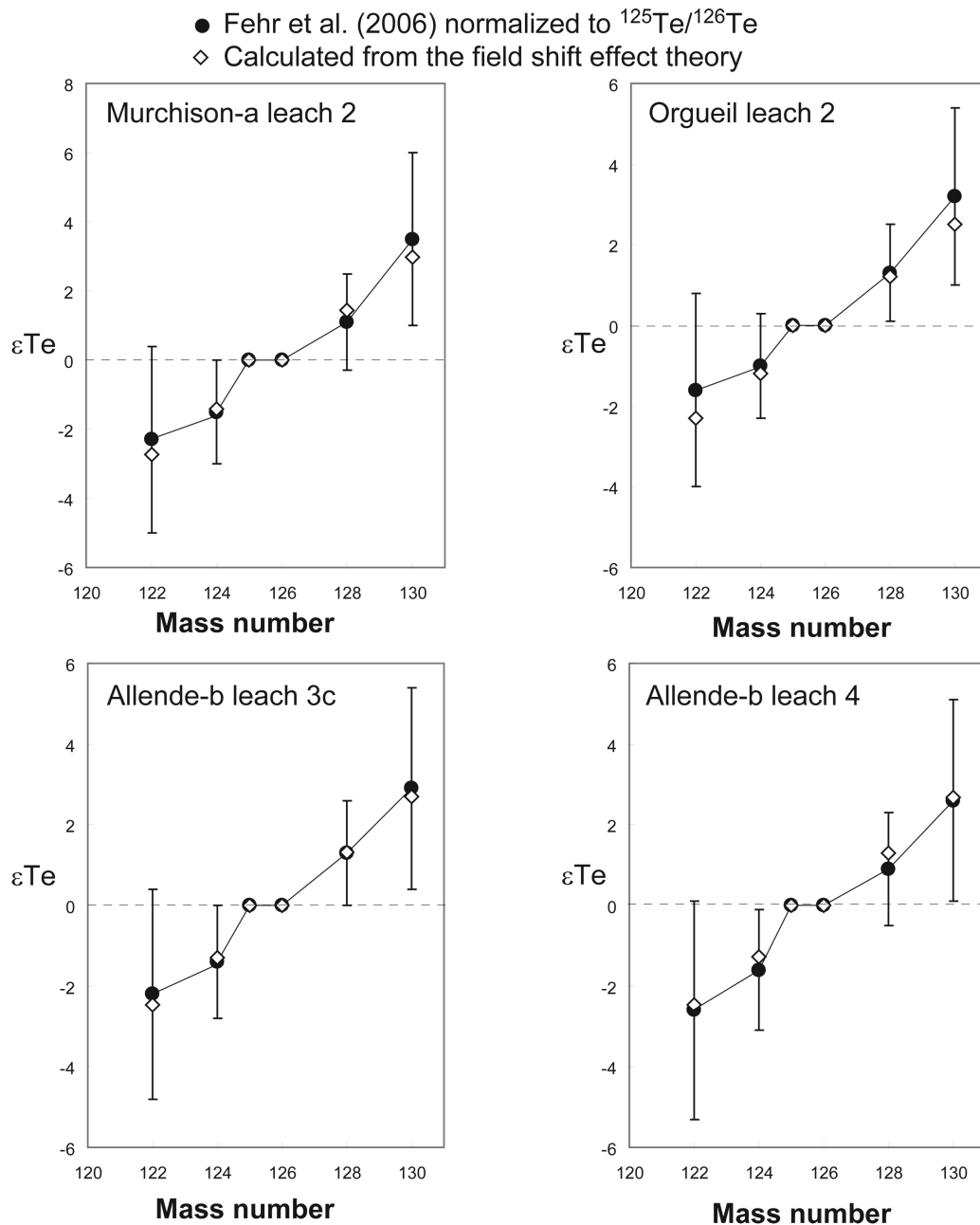


Fig. 3. Isotopic variations of Te in leachates from Allende, Murchison, and Orgueil (Fehr et al. 2006) normalized to $^{125}\text{Te}/^{126}\text{Te} = 0.374902$ (Lee and Halliday 1995). ^{120}Te data are not shown due to their large error. The calculated symbols correspond to the fit of the measured data by the nuclear field shift theory using Equation 2.

analytical artifacts: first, because Fehr et al. (2006) performed extensive matrix tests including natural samples and synthetic standard solutions to check that their ^{130}Te anomalies were real. Second, if a simple acid leaching could create such remarkable patterns due to field shift effect, most metamorphic samples (e.g., the LL6 chondrite Allan Hills [ALH] 84081 of Fehr et al. 2005) would also be isotopically anomalous. We contend, instead, that a ratio of isotopes of different parity, here $^{125}\text{Te}/^{126}\text{Te}$, is most susceptible to non-mass dependent effects and thus using such a ratio for

normalization may affect the entire abundance spectrum with the heavier isotopes, here ^{130}Te , giving the spurious impression of strong anomalies. In order to demonstrate that the $^{125}\text{Te}/^{126}\text{Te}$ ratio does not provide optimal normalization, we first computed a histogram of Pearson correlation coefficients ρ between the mass number and ϵ values for Murchison-a leach 2 data (Fehr et al. 2006) drawing random deviates of the observed values in a Monte Carlo experiment (Fig. 4). The values of correlation coefficients obtained from Fehr et al.'s (2006) data normalized to $^{125}\text{Te}/^{126}\text{Te}$ have a

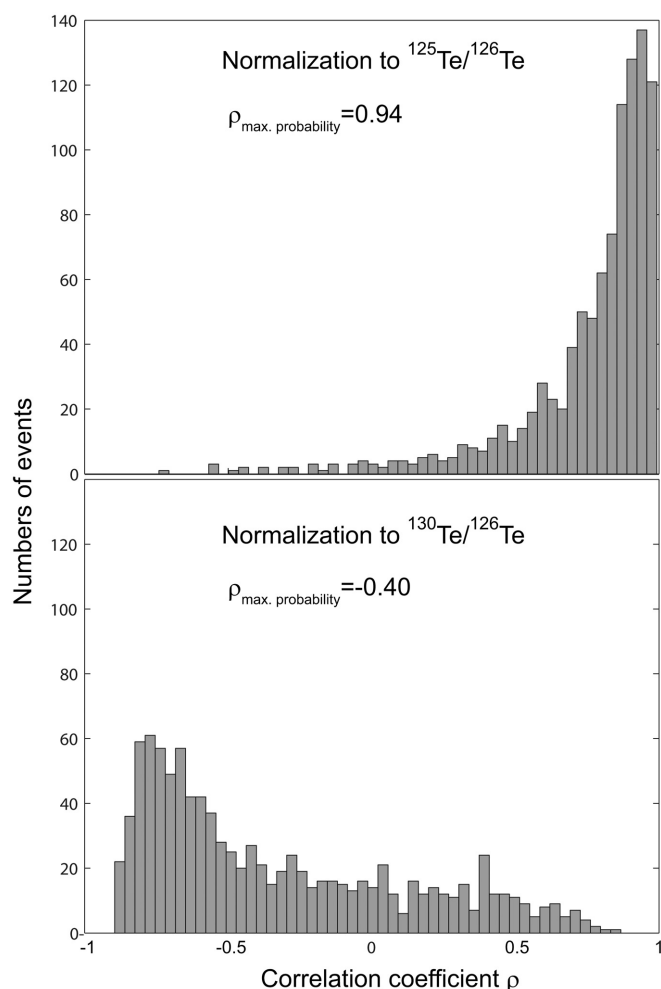


Fig. 4. Histograms of Pearson's correlation coefficient between ϵ_{Te} and the masses of the isotopes for Murchison-a leach 2 (Fehr et al. 2006). The errors on the isotopic ratios are propagated using a Monte Carlo simulation with 1000 events. $\rho_{\text{max, probability}}$ refers to the correlation coefficient with the maximum of probability.

maximum-likelihood ρ_{max} value of 0.94, which indicates a strong residual effect of mass-dependent fractionation. The histogram of Fig. 4 also shows that the probability that $\rho_{\text{max}} = 0$ is essentially negligible. In comparison, once normalized to $^{130}\text{Te}/^{126}\text{Te} = 1.81684$ Fehr et al. (2005), ρ_{max} drops down to -0.40 . Although the latter value is still significant, the spread of the histogram shows that the probability that $\rho_{\text{max}} = 0$ (no residual correlation with mass after normalization) is now very substantial. This clearly shows that normalization to $^{125}\text{Te}/^{126}\text{Te}$ leaves a residual correlation with mass number much larger than normalization to $^{130}\text{Te}/^{126}\text{Te}$. We therefore use an alternative normalization to the $^{130}\text{Te}/^{126}\text{Te}$ ratio, two even isotopes, whose higher abundances minimize error propagation. This normalization leaves only the ^{125}Te abundance as clearly anomalous (Fig. 5). Because Ag concentrations upon chemical purification are too low to create interferences at the level requested to explain the

anomalies observed in their leachates, small interferences with $^{109}\text{Ag}^{16}\text{O}$ at mass 125 by checking mass 109 can be safely ruled out (Fehr et al. 2005).

The odd ^{125}Te nuclide contains variable contributions from *s* and *r* processes, but so does ^{126}Te (Arlandini et al. 1999). In contrast the ^{128}Te and ^{130}Te are pure *r*-process nuclide. Therefore, an isolated deficit or an excess of ^{125}Te is unlikely to result from a different contribution of one of the processes.

By adjusting the parameter *a* from Equation 2, we show in Figs. 3 and 5 that the Te isotope pattern can be exactly accounted for by the nuclear field shift theory for each normalization (to $^{125}\text{Te}/^{126}\text{Te}$ and $^{130}\text{Te}/^{126}\text{Te}$).

As for Mo and Ru in Murchison (Fujii et al. 2006b), the processes that created mass-independent fractionation effects and their site in the planetary nebula are unknown. Since essentially little is known about the mass-dependent fractionation of Te isotopes (a common situation for heavy elements) with the exception of the pioneering work of Fehr et al. (2005), it is difficult to be more specific for mass-independent fractionation processes. It is, however, likely that they both occur simultaneously in the same environment, but that only the mass-independent part of isotopic fractionation remains after normalization to a reference ratio. It can be speculated that Te isotope anomalies arise during a phase change, e.g., volatilization in the nebular environment or metamorphism on the parent body of the carbonaceous chondrites, rather than through nucleosynthetic processes. Te is a volatile element, chemically similar to S and Se, and has three oxidation states, Te(II), Te(IV), and Te(VI). It can therefore be expected that Te forms solid solutions with sulfides (Cotton and Wilkinson 1988). A first possibility based on the volatility of Te (50% condensation temperature of 709 K [Lodders 2003]) is isotope fractionation during the interaction of solid phases with the nebular gas during the early stages of condensation. Congruent evaporation of forsterite (Hashimoto 1990) and incongruent evaporation of enstatite (Tachibana et al. 2002), melt (Wang et al. 2001) and silicate glass (Xue et al. 1996) further are known to result in the isotopic fractionation of various metals (Si, Mg, Zn). Although the experiments attempting to reproduce the reverse process of condensation are not as well understood (Richter 2004), isotopic fractionation is likely to accompany the formation of mineral and metal grains in the nebula.

Alternatively, redox reactions between the dominant Te anionic species present in metamorphic fluids and interaction of these fluids with sulfides on the parent bodies of meteorites are likely to create both mass-dependent and mass-independent isotopic fractionation. Some of the sulfides present in carbonaceous chondrites may indeed have formed by sulfidation of FeNi metal in the meteorite parent body (Boctor et al. 2002). We speculate, however, that the general lack of mass-independent effects in bulk meteoritic material (Fehr et al. 2005); in contrast with their presence in the leaching of rather primitive carbonaceous chondrites suggests

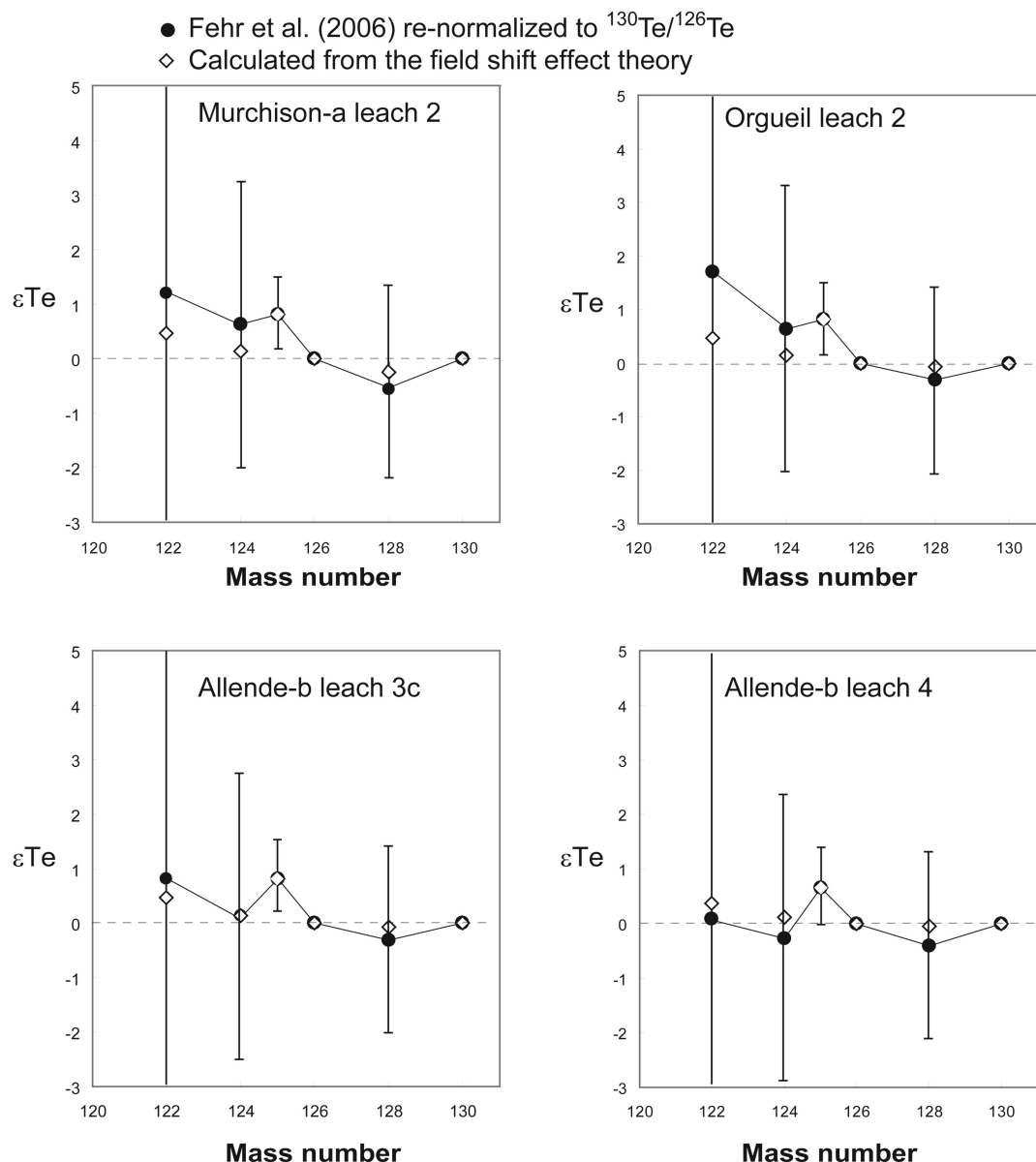


Fig. 5. Isotopic variations of Te in leachates from Allende, Murchison, and Orgueil (Fehr et al. 2006) normalized to $^{130}\text{Te}/^{126}\text{Te} = 1.81684$ (Fehr et al. 2005), which is the value of the unprocessed Te standard solution. The calculated symbols correspond to the fit of the measured data by the nuclear field shift theory using Equation 2.

that mass-independent isotopic patterns are hosted in matrix sulfides precipitated from the nebular gas and do not arise from late-stage metamorphic processes.

Whether mass-independent effects fingerprint high-temperature fractionation between the nebular gas and its condensates or fractionation at low temperature during interaction between silicates and fluids on the parent body remains to be elucidated. A guideline for interpretation may be found in Bigeleisen's theory, which suggests that at high temperature, the effect of field shift (in $1/T$) dominates mass-dependent effects (in $1/T^2$). In contrast to the mass-dependent effect, which may often be predicted by the stiffness of molecular bonds, the direction of mass-independent

fractionation in a particular mineral, and consequently its relative excess/deficit of individual nuclides, cannot be easily predicted and may change with the extent of the reaction. We suggest, however, that a wealth of information is hidden in mass-independent fractionation effects.

CONCLUSIONS

We propose that the Te isotopic anomalies in 2 bulk CAIs from Allende (Fehr et al. 2009) and in leachates of Allende, Murchison, and Orgueil (Fehr et al. 2006) may be explained by mass-independent effects resulting from either metamorphic processes on the meteorite parent body or,

preferably, interactions of condensed solids with the nebular gas. Field shift effect is thus an alternative origin to explain anomalous Te isotopic compositions.

Acknowledgments—This work was supported in part by a NASA grant NNX09AM64G (FM). We received many helpful comments from two anonymous reviewers, the editor, and the associate editor. Sune Nielsen, Andrew Davis, one anonymous reviewer and Sara Russell are also thanked for comments on a previous version of this paper. We thank Janne Blichert-Toft for careful English editing.

Editorial Handling—Dr. Christine Floss

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