

## Geochemical constraints on the half-life of $^{130}\text{Te}$

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To determine the half-life of  $^{130}\text{Te}$  we have analyzed multiple aliquots of geological telluride samples 100 times smaller than those previously reported using a unique resonance ionization mass spectrometer. We employ a low-fluence neutron irradiation that allows determination of parent and daughter from the same xenon isotopic analysis. Step heating of these irradiated samples allows the  $^{130}\text{Xe}/^{132}\text{Xe}$  ratio of fluids trapped inside the tellurides to be determined. Considering only samples where the trapped  $^{130}\text{Xe}/^{132}\text{Xe}$  ratio is demonstrably consistent with atmospheric xenon, we can avoid over- or under-estimating the half-life due to redistribution or inheritance of radiogenic  $^{130}\text{Xe}$ . Combining our work with literature data, it is clear that several relatively young samples have retained xenon quantitatively since formation, allowing the half-life to be determined as  $(8.0 \pm 1.1) \times 10^{20}$  yr. Older samples have clearly been affected by post-formation processing. This suggests that there is little hope of monitoring solar luminosity through the geological record of  $^{126}\text{Xe}$  production by solar neutrinos, but it is possible that geologically useful chronological information can be obtained from this system.

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### I. INTRODUCTION

The half-life of  $^{130}\text{Te}$  against neutrinoless  $\beta\beta$ -decay has been shown to be  $>1.8 \times 10^{24}$  yr using a calorimetric technique [1]. In the same experiment, the half-life of  $^{130}\text{Te}$  for lepton-conserving double beta decay has been reported as being in the range  $(1.2\text{--}10.4) \times 10^{20}$  yr (incorporating systematic and statistical errors) [2]. Double beta decay of this isotope was first demonstrated by Inghram and Reynolds [3], who studied tellurium-rich geological samples and reported a half-life of  $1.4 \times 10^{21}$  yr. Since this first attempt, multiple analyses of telluride minerals have been attempted, with half-lives being reported in the range from  $(7.0\text{--}27) \times 10^{20}$  years [4,5]

Several effects may have contributed to this range in previously reported half-lives. The half-life will be underestimated if a telluride mineral inherited  $^{130}\text{Xe}$  on formation that had been produced in a previous generation of tellurides. It will be overestimated if some of the  $^{130}\text{Xe}$  generated in the sample since formation has been lost. The observed range indicates that one or both of these processes is prevalent, so both must be excluded before confidence can be placed in a result from any sample.

Measuring the absolute half-life of  $^{130}\text{Te}$  geochemically depends on measuring the amount of  $^{130}\text{Xe}$  (the daughter product) that has been produced over a known time from a known amount of tellurium in a mineral. It is thus a requirement that xenon has been retained quantitatively since a well defined event of known age either the original mineral formation or a subsequent significant geothermal event. If this condition is satisfied, in addition to the half-life determination,  $^{126}\text{Xe}$  produced by interaction of solar neutrinos with  $^{126}\text{Te}$  might test the predicted increase in solar luminosity over geological time

(should a suitable sample be identified) [6] and  $\beta\beta$ -decay of  $^{130}\text{Te}$  might provide a useful method of dating mineral deposits of economic importance. For the latter application, and for half-life determination, it is also necessary that the amount of the daughter isotope present can be unambiguously divided between any trapped on formation and that subsequently produced *in situ* by  $^{130}\text{Te}$  decay.

Telluride minerals can exhibit varying stoichiometry, and are typically minor phases in a mineralising event which are hard to separate from other co-precipitated minerals. The sensitivity of conventional noble gas mass spectrometry is such that samples larger than those that typically form naturally (several hundred milligrams or more) are required, further restricting the useful sample set. Literature studies have thus been restricted to rare massive samples of tellurides with well defined compositions. The most common occurrence of telluride minerals is in hydrothermal vein-gold mineralisation where establishing the age of mineralizing relies on isotopic geochronometers, thus establishing the half-life of  $^{130}\text{Te}$  and a microanalysis system provides the potential of a new geochrometer.

We have overcome these constraints in two ways. Use of the resonance ionization instrument RELAX (Refrigerator Enhanced Laser Analyser for Xenon [7]), the state-of-the-art in xenon isotopic analysis, has enabled samples smaller than 1 mg to be studied. Also, our samples were subjected to a low-fluence neutron irradiation before analysis, converting  $^{130}\text{Te}$  to  $^{131}\text{Xe}$  via  $^{131}\text{Te}$  and  $^{131}\text{I}$  [8]. This allows the tellurium content of samples to be measured as part of the same xenon isotopic analysis as the concentration of its daughter product, removing the need for samples of pure minerals with well defined stoichiometry. Telluride minerals such as those used to measure the half-life were deposited alongside a range of other minerals from aqueous fluids that had moved through a volume of the Earth's crust. When combined with step heating, analysis of irradiated samples also allows the  $^{130}\text{Xe}/^{132}\text{Xe}$  of xenon associated with residual parent fluids trapped in the mineral (rather than correlated with tellurium) to be determined. This

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can reveal both inheritance of radiogenic  $^{130}\text{Xe}$  on formation of the mineral, and redistribution/loss of radiogenic  $^{130}\text{Xe}$  from its formation site, as discussed below.

Here, the development and testing of this technique is described. We investigate to what extent the requirement is met that xenon is not inherited and is retained, and discuss implications for geochemical half-life determinations and for the use of  $^{130}\text{Te}$  decay as a chronometer of ore deposition.

## II. METHODS

The range of reported half-lives corresponds to  $^{130}\text{Xe}/^{130}\text{Te} = (2.5\text{--}10) \times 10^{-13}$  for minerals that have retained xenon for 1 Gyr. RELAX is a time-of-flight mass spectrometer, so all isotopes are measured on the same (microchannel plate) detector. The dynamic range of the detector means that optimum performance will be achieved when  $^{131}\text{Xe}/\text{Te}$  after irradiation is within a factor of  $\sim 10$  of  $^{130}\text{Xe}/\text{Te}$  in the sample. There is, however, a need to take account of  $^{131}\text{Xe}$  produced by interaction of the sample with neutrons in the crust over geological periods. Minimizing the vulnerability of our technique to this correction favors higher fluences so that reactor production dominates this component. The cross section for radiative capture of thermal neutrons of  $^{130}\text{Te}$  is 270 mb, so we adopted nominal fluences of  $10^{13} \text{ n cm}^{-2}$  for our irradiations. (A reviewer points out that since these experiments were planned the neutron capture cross section of  $^{130}\text{Te}$  has been revised downwards by  $\sim 20\%$  [9]. This change has no effect on the results presented here since the tellurium-xenon conversion efficiencies of our irradiations were measured rather than calculated—the cross section was used only in targeting a desirable neutron fluence.) Irradiations were performed using the Automatic Large Volume Irradiation System (ALVIS) at the Imperial College Reactor Centre, with 2 cm diameter pots capable of holding 6–7 separate aliquots. Irradiated aliquots were retained for three months before analysis to allow production of  $^{131}\text{Xe}$  via  $^{131}\text{I}$  (half-life 8.02 d). Not all irradiated aliquots were analyzed.

Aliquots of native tellurium from Good Hope were included in every irradiation to monitor conversion efficiency ( $^{131}\text{Xe}^{**}/\text{Te}$  for an irradiation, where  $^{131}\text{Xe}^{**}$  is the  $^{131}\text{Xe}$  produced in the irradiation from  $^{130}\text{Te}$ ).  $^{130}\text{Xe}^*/^{131}\text{Xe}^{**}$  ratios (where  $^{130}\text{Xe}^*$  is  $^{130}\text{Xe}$  produced by  $\beta\beta$ -decay of  $^{130}\text{Te}$ ) in aliquots of other samples were then determined as multiples of that in Good Hope. The precision with which this relative ratio can be determined is limited by the variation in conversion efficiency (variation in neutron fluence) across an irradiation pot and variation in  $^{130}\text{Xe}^*/\text{Te}$  among aliquots of the Good Hope sample. Precision was evaluated using irradiations with multiple Good Hope aliquots.

Relative  $^{130}\text{Xe}^*/^{131}\text{Xe}^{**}$  ratios are equivalent to relative  $^{130}\text{Xe}^*/\text{Te}$  ratios. For geologically relevant periods,  $^{130}\text{Xe}^*/\text{Te}$  increases linearly with time, so relative  $^{130}\text{Xe}^*/\text{Te}$  ratios correspond to relative  $^{130}\text{Xe}$  retention ages. These can be directly compared to relative dates of geological events associated with the samples' source areas, allowing the hypothesis of quantitative retention to be examined. Constraining the half-life additionally requires an absolute determination of

$^{130}\text{Xe}^*/^{130}\text{Te}$  for one sample and identification of an absolute age for one sample.

Xenon has nine stable isotopes, but our data analysis involves only those with masses 130, 131, 132, 134, and 136. Of these,  $^{130}\text{Xe}$  and  $^{131}\text{Xe}$  are tellurogenic (produced from tellurium);  $^{130}\text{Xe}$  is not produced in the reactor, but  $^{131}\text{Xe}$  production occurs by neutron capture both in the crust and in the reactor.  $^{131\text{--}136}\text{Xe}$  are produced in known ratios by spontaneous fission of  $^{238}\text{U}$ , and all five are present in air. To calculate  $^{130}\text{Xe}^*/^{131}\text{Xe}^{**}$  it is necessary to disentangle the contributions from these various sources.

Any  $^{132}\text{Xe}$ ,  $^{134}\text{Xe}$ , and  $^{136}\text{Xe}$  present will be a mixture of air and fission xenon. If the  $^{136}\text{Xe}/^{132}\text{Xe}$  ratio of fission xenon is  $R_F$ , that of air xenon is  $R_A$  and that measured is  $R_M$  then the fraction of  $^{132}\text{Xe}$  contributed by air xenon is  $f_A = (R_M - R_F)/(R_A - R_F)$ . Once the amounts of  $^{132}\text{Xe}$  contributed from fission of  $^{238}\text{U}$  and from air have been calculated, the  $^{131}\text{Xe}/^{132}\text{Xe}$  ratio corresponding to the mixture of trapped and fission xenon present can be determined and  $^{131}\text{Xe}^*/^{130}\text{Xe}$  calculated as the excess in the measured ratio over this quantity. In practice, few samples showed evidence of fission xenon, but hereafter  $^{132}\text{Xe}$  refers exclusively to the fission corrected quantity.

After correcting for fission, xenon released from samples will be a mixture of xenon associated with the trapped fluid (with  $^{131}\text{Xe}^*/^{130}\text{Xe} = 0$ ) and tellurogenic xenon produced *in situ* (with  $^{132}\text{Xe}/^{130}\text{Xe} = 0$ ). Separate analyses of a single aliquot are expected to exhibit different proportions of trapped and tellurogenic xenon and, if our expectation is correct, will form linear arrays on graphs of  $^{132}\text{Xe}/^{130}\text{Xe}$  vs  $^{131}\text{Xe}^*/^{130}\text{Xe}$ .

To test the mixing hypothesis a stepwise heating analytical procedure was adopted. Irradiated aliquots were loaded into a port of the RELAX mass spectrometer still wrapped in the Al foil in which they had been irradiated. They were heated for one minute at each of a series of sequentially increasing powers by a continuous wave Nd:YAG (1064 nm) laser. At each power setting evolved gas was gettered for 1 min to remove active gases then admitted to the mass spectrometer and analyzed. Residual gas was then pumped away in preparation for analysis at the next power setting. In graphs such as Fig. 1, where data from unirradiated and irradiated aliquots of the same sample are plotted and exhibit separate correlations, the intercept with the horizontal axis corresponds to the ratio of  $^{131}\text{Xe}^*/^{130}\text{Xe}$  for the tellurogenic component (i.e., no  $^{132}\text{Xe}$  is present), which is equal to  $^{131}\text{Xe}^*/^{130}\text{Xe}^*$ . The intercept with the vertical axis corresponds to the  $^{132}\text{Xe}/^{130}\text{Xe}$  ratio of the trapped component (i.e., no  $^{131}\text{Xe}^*$  is present). Of the samples that showed evidence of a trapped component (some yielded no  $^{132}\text{Xe}$  or produced no correlation), only Kalgoorlie showed conclusive evidence of the presence of radiogenic  $^{130}\text{Xe}$  (Fig. 2).

In order to determine  $^{131}\text{Xe}^{**}/^{130}\text{Xe}^*$  for the tellurogenic component, similar step heating experiments were performed on duplicate samples that had not been subject to artificial irradiation, where

$$\frac{^{131}\text{Xe}^{**}}{^{130}\text{Xe}^*} = \frac{^{131}\text{Xe}^*}{^{130}\text{Xe}^*_{\text{irr}}} - \frac{^{131}\text{Xe}^*}{^{130}\text{Xe}^*_{\text{unirr}}} \quad (1)$$

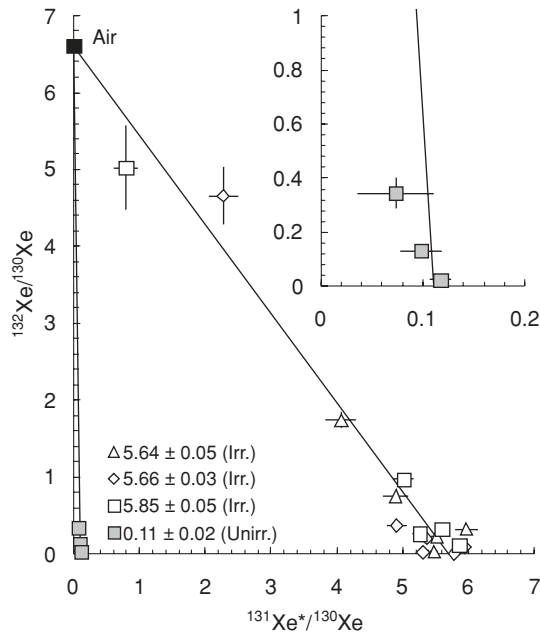


FIG. 1. Fission corrected data from three aliquots of native tellurium from Good Hope included in irradiation Ascot 2 (Irr., Table I) and one unirradiated aliquot (Unirr., inset). Data lie close to mixing lines (shown) between the tellurogenic components calculated from Williamson fits and trapped xenon, in this case the latter is consistent with air-derived gas. Reactor produced  $^{131}\text{Xe}^*$  dominates that produced by the crustal neutron fluence by a factor of 50. Data sets are labeled with  $^{131}\text{Xe}^*/^{130}\text{Xe}^*$  values determined from Williamson fits and derived  $1\sigma$  errors. Scatter among the irradiated samples corresponds to  $\pm 2\%$  variation in  $^{131}\text{Xe}^*/^{130}\text{Xe}^*$ .

The subscripts *irr* and *unirr* denote ratios derived from irradiated and unirradiated samples respectively, and other symbols are defined above.

### III. RESULTS

Data from the seven separate irradiations discussed here are summarized in Table I, complete data tables are available from the correspondence author.

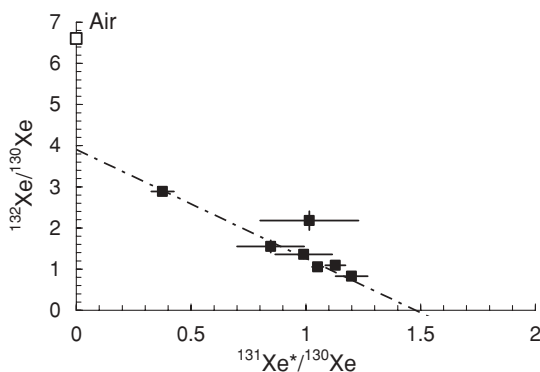


FIG. 2. Data from a sample of Kalgoorlie melonite ( $\text{NiTe}_2$ ) reveal a trapped component with a lower  $^{132}\text{Xe}^*/^{130}\text{Xe}^*$  ratio than air. 23% of the  $^{130}\text{Xe}^*$  in this sample is not correlated with tellurium.

TABLE I. In the first column,  $^{130}\text{Xe}^*/^{131}\text{Xe}^{**}$  and sample mass is given for each aliquot of native tellurium from Good Hope in each irradiation, and the adopted value for the irradiation shown in bold. Other samples included in the same irradiation are named in the second column, and their  $^{130}\text{Xe}^*/\text{Te}$  ratios are given as fractions of the adopted Good Hope ratio in the third column: S. C. = Southern Cross.

$^{130}\text{Xe}^*/^{131}\text{Xe}^{**}$ in Good Hope, Mass <sup>a</sup>	Sample	$^{130}\text{Xe}^*/\text{Te}$ , Mass
<b>Ascot 1</b>		
$0.181 \pm 0.002$ , 1.0 mg	Kochbulak	$0.55 \pm 0.01$ , 1.6 mg
$0.187 \pm 0.001$ , 2.1 mg	Clogau	$0.60 \pm 0.01$ , 1.6 mg
$0.182 \pm 0.001$ , 1.2 mg	Clogau	$0.64 \pm 0.02$ , 1.1 mg
<b><math>0.184 \pm 0.003</math></b>	Clogau	$0.57 \pm 0.02$ , 2.9 mg
<b>Ascot 2</b>		
$0.181 \pm 0.002$ , 1.7 mg	Kochbulak	$0.56 \pm 0.02$ , 1.2 mg
$0.180 \pm 0.002$ , 1.6 mg		
$0.175 \pm 0.002$ , 1.3 mg		
<b><math>0.179 \pm 0.004</math></b>		
<b>Ascot 3</b>		
$0.169 \pm 0.002$ , 0.4 mg	Kochbulak <sup>b</sup>	$0.53 \pm 0.09$ , 2.6 mg
<b><math>0.169 \pm 0.004</math></b>	Kochbulak <sup>c</sup>	$0.60 \pm 0.02$ , 2.2 mg
	Kochbulak	$0.58 \pm 0.02$ , 0.4 mg
<b>Ascot 4</b>		
$0.225 \pm 0.004$ , 0.4 mg	Kochbulak	$0.59 \pm 0.01$ , 0.5 mg
<b><math>0.225 \pm 0.004</math></b>	S. C. melonite	$3.01 \pm 0.10$ , 1.3 mg
	Kalg. melonite	$0.02 \pm 0.00$ , 1.3 mg
<b>Ascot 7</b>		
$0.212 \pm 0.009$ , 0.9 mg	Boliden	$1.63 \pm 0.15$ , 0.7 mg
<b><math>0.212 \pm 0.009</math></b>		
<b>Ascot 17</b>		
<b><math>0.323 \pm 0.009^d</math></b>	Kochbulaklak	$\approx 0.59 \pm 0.01$ , 0.9 mg
	Kalg. Krennerite	$1.65 \pm 0.05$ , 1.0 mg
	Kalg. Calaverite	$2.23 \pm 0.07$ , 1.1 mg
<b>Ascot 18</b>		
$0.165 \pm 0.001$ , 0.8 mg	Kochbulak	$0.66 \pm 0.02$ , 0.9 mg
$0.157 \pm 0.001$ , 0.7 mg	Kochbulak	$0.58 \pm 0.02$ , 0.9 mg
<b><math>0.161 \pm 0.004</math></b>	Kochbulak	$0.61 \pm 0.02$ , 0.7 mg

<sup>a</sup>Errors on adopted ratios cover the range of individual analyses or reflect a typical range when only one Good Hope sample is reported.

<sup>b</sup>Distinct sample (altaite).

<sup>c</sup>Distinct sample (native tellurium).

<sup>d</sup>No Good Hope sample available. Adopted value calculated from the Kochbulak native tellurium sample assuming the average fractional  $^{130}\text{Xe}^*/\text{Te}$  of Kochbulak relative to Good Hope determined for other irradiations.

In Fig. 1 we evaluate the limit on precision of our technique imposed by variations in neutron fluence and in  $^{130}\text{Xe}^*/\text{Te}$  of our monitor using data from three aliquots of native tellurium from Good Hope included in the Ascot 2 irradiation. Data show the expected variable mixtures of terrestrial air and tellurogenic xenon, the unirradiated sample having  $^{131}\text{Xe}^*/^{130}\text{Xe}^*$  lower by a factor of 50. Variations in the  $^{131}\text{Xe}^*/^{130}\text{Xe}^*$  ratio of

the irradiated samples amount to  $\pm 2\%$ . Since this was the irradiation which exhibited most variability among multiple Good Hope samples, the spread in the ratios derived from this irradiation were adopted as the limit on precision for determining the  $^{131}\text{Xe}^*/^{130}\text{Te}$  conversion factor for irradiations where multiple aliquots of the monitor did not allow this to be assessed independently.

The  $^{130}\text{Xe}^*/^{131}\text{Xe}^{**}$  ratio of native tellurium from Kochbulak relative to the Good Hope sample was measured in five separate irradiations.  $^{130}\text{Xe}^*/^{131}\text{Xe}^{**}$  in Good Hope varied by a factor of 2 across these irradiations, while the ratio of Kochbulak relative to co-irradiated Good Hope was  $0.579 \pm 0.026$  (mean, standard deviation), a precision of 4.4% which is consistent with derived uncertainties.

Absolute amounts of  $^{130}\text{Xe}^*$  in Good Hope native tellurium were determined by mixing gas released from the sample with aliquots of a monoisotopic spike consisting of known amounts of  $^{128}\text{Xe}$  [7]. Derived gas quantities varied by a factor of 2.5, more than expected from derived uncertainties, which we attribute to incomplete degassing of some of the samples within their Al wraps. Of the ten samples analyzed, five were consistent with a  $^{130}\text{Xe}^*/\text{Te}$  ratio of  $(7.47 \pm .35) \times 10^8$  atoms  $\text{g}^{-1}$ , while the others suggested lower ratios consistent with incomplete degassing. This corresponds to an atomic  $^{130}\text{Xe}/^{130}\text{Te}$  ratio of  $(4.79 \pm 0.22) \times 10^{-13}$ , which we adopt as representative of our Good Hope sample. Literature data for what we believe to have been other portions of the same sample correspond to:  $(4.72 \pm 0.29) \times 10^{-13}$  [8],  $(4.01 \pm 0.20) \times 10^{-13}$  [10],  $(4.12 \pm 0.69) \times 10^{-13}$  [11], and  $(3.62 \pm 0.14) \times 10^{-13}$  [12].

#### IV. COMPARISON WITH GEOLOGICAL DATES

The orebodies from which the telluride minerals are sourced were deposited from fluids into pre-existing rock (so-called ‘epigenetic deposits’) and thus these surrounding rocks may not be genetically related to the mineralizing episode and the age of their formation can predate it by hundreds of millions of years. There are relatively few isotopic dating systems applicable to ore minerals themselves in these orebodies and it remains to be demonstrated that the Te-Xe system reflects the mineralizing event. As, there may have been more than one episode of ore deposition in any locale, so identifying and dating a candidate geological event to associate with the start of xenon retention in a telluride is not trivial. Furthermore, post formation processing of deposits may result in partial loss of xenon that has already been generated, in which case  $^{130}\text{Xe}^*/\text{Te}$  will not correspond to a meaningful age. Such a process can explain the Kalgoorlie data of Fig. 2—the  $^{130}\text{Xe}^*$  in the trapped component was redistributed into fluid inclusions from adjacent telluride material. Such a process cannot be assumed to preserve bulk  $^{130}\text{Xe}^*/\text{Te}$ , nor can it be assumed to have completely removed all  $^{130}\text{Xe}^*$  present when the event occurred from correlation with its parent tellurium, thus the derived age is unlikely to match the date of a geological process. We suggest that an important constraint on the reliability of a half-life determination from a telluride mineral is that there be firm evidence of an accompanying trapped component that has not been modified

by redistribution of  $^{130}\text{Xe}$ . Adopting this constraint insures against underestimating the half-life from a sample that has in fact lost tellurogenic xenon. Takaoka *et al.* [13] measured fluids in co-precipitated quartz and arsenopyrite to ensure no  $^{130}\text{Xe}^*$  was present in the formation fluid, but such measurements will not detect redistribution of  $^{130}\text{Xe}^*$  within the telluride and associated loss.

We now discuss the geological context of our sample suite, seeking to constrain dates for the onset of xenon retention.

Kochbulak is in the Kurama range, Central Tien Shan, Uzbekistan. Ore deposition is associated with production of a potassium bearing mica amenable to K-Ar dating, which has resulted in a relatively well constrained age of 260–285 Myr [14]. At some point after deposition the deposit may have been affected by the event that resulted in the formation of quartz-carbonate-barite veinlets [14]. We have analyzed multiple aliquots of the same native tellurium sample, a second native tellurium sample sourced from a different collection, and a sample of altaite (PbTe) from this deposit.

Clogau is part of the Dolgellau gold belt in the United Kingdom. K-Ar dating of micas associated with post-depositional geological activity yields ages around 400 Myr [15], while the minerals themselves are thought to have been deposited at 476–493 Myr [16]. Our tetradymite ( $\text{Bi}_2\text{Te}_2\text{S}$ ) samples are sourced from two well separated locations, where they were present as a superficial veneer on country rock, from which only partial separation was possible.

The Good Hope goldmine is located in Colorado (USA). Two episodes of mineralization have occurred, the second of which is responsible for telluride deposition [17], while separate episodes of igneous activity occurred in the vicinity and have been dated to 564–573 Myr and to the last 30 Myr [18]. Pb-Pb analyses of the tellurides yielded an age of 1.3 Gyr [19], but this technique depends on the model of Pb isotopic evolution via uranium decay in the source region that is adopted. In this case, the presence of a generation of ore deposition that predates telluride formation makes simple interpretation of the Pb system problematic, but the system may be seen as providing an upper limit to the age of the deposit. We adopt the dated igneous activity as a lower limit, though cannot rule out more recent deposition. We have analysed multiple aliquots of the same sample of native tellurium from Good Hope.

The Boliden deposits are situated in the Fennoscandian shield in Northern Sweden. Geological studies suggest original deposition and subsequent remobilization at 1.89–1.85 Gyr [20]. A U-Xe age of  $1.56 \pm 0.39$  Gyr has been reported while U-He and K-Ar data suggest an age of  $540 \pm 10$  Myr [21]. Our sample is the mineral tellurobismuthite,  $\text{Bi}_2\text{Te}_3$ .

Kalgoorlie and Southern Cross are hosted in the Yilgarn block of Western Australia and mineralization has been established as having occurred at 2.62–2.64 Gyr [22] with some igneous activity 20–30 Myr later and little evidence of disturbance since this time [23]. We have analyzed Krennerite ( $\text{NiTe}_2$ ), Melonite ( $\text{AuTe}_2$ ), and Calaverite ( $\text{AuTe}_2$ ) from Kalgoorlie and melonite from Southern Cross.

In Fig. 3  $^{130}\text{Xe}^*/\text{Te}$  ratios relative to that in Good Hope are compared with the relative ages from geological dating. If all samples had retained xenon quantitatively from a well

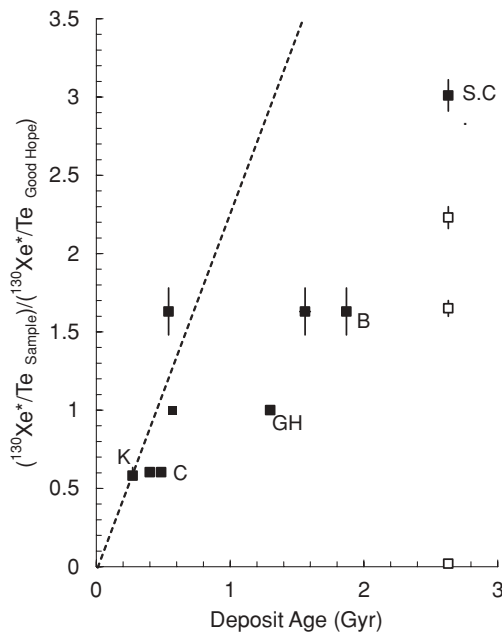


FIG. 3. Comparison of relative  $^{130}\text{Xe}^*/\text{Te}$  values with chronological data for the relevant deposits. K—Kochbulak, C—Clogau, GH—Good Hope, B—Boliden, S. C.—Southern Cross. Kalgoorlie samples are shown with open symbols. Horizontal tie lines join data where different geological ages are possible for the same sample. The dashed line represents the trend expected for perfect xenon retention from the accepted formation ages if the Kochbulak datum represents an accurate Te-Xe formation age.

defined age, we would expect a linear correlation. However, the observation of an evolved trapped component in Kalgoorlie (Fig. 2) is consistent with the redistribution of radiogenic  $^{130}\text{Xe}$  that would account for Te-Xe ages being shorter than the accepted formation age. Figure 3 suggests that an upper limit to the half-life of  $^{130}\text{Te}$  from these data should be based either on Kochbulak or Boliden. Of these, we can eliminate the possibility that the Kochbulak telluride significantly predates the accepted age, since this deposit is part of widescale mineralization associated with a continental margin volcanic belt, formed in the Late Palaeozoic on a Precambrian and Early Hercynian continental basement [24]. In contrast, the Boliden telluride was definitely present for at least 1 billion years before the U-He and K-Ar ages. We thus identify the  $^{130}\text{Xe}^*$  content of Kochbulak native tellurium samples from two different sources and altaite as providing the most stringent upper limit on the half-life of  $^{130}\text{Te}$ .

The relative  $^{130}\text{Xe}^*/\text{Te}$  value for Kochbulak native tellurium combined with the  $^{130}\text{Xe}^*/^{130}\text{Te}$  ratio of Good Hope yields an atomic  $^{130}\text{Xe}^*/^{130}\text{Te}$  ratio of  $(2.79 \pm 0.28) \times 10^{-13}$ . Adopting the age restraint above for this deposit gives what is strictly an upper limit on the  $^{130}\text{Te}$  half-life of  $(6.8 \pm 0.8) \times 10^{20}$  yr based on the samples reported here since, if Kochbulak tellurides have been reset since formation of the deposit, the half-life of  $^{130}\text{Te}$  is shorter than this. Against this possibility are the observations that multiple samples yield the same half-life, and that the fluid phase trapped in the telluride shows no evidence of  $^{130}\text{Xe}^*$ .

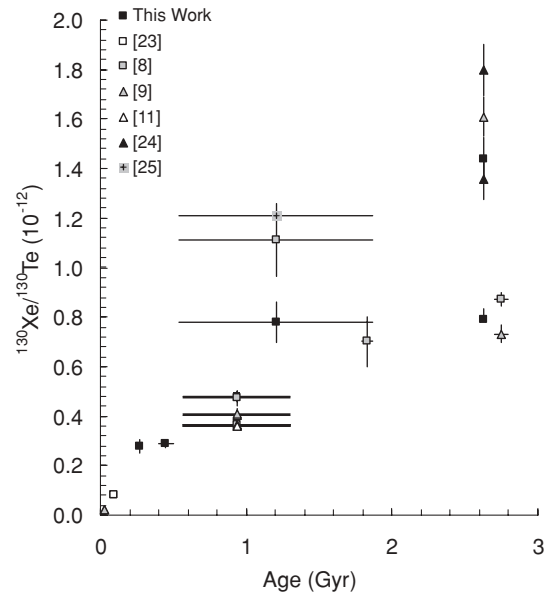


FIG. 4. Comparison of our data with that in the literature. Data from a single deposit lie in vertical lines, while horizontal lines reflect the range of plausible ages for deposits. Data from the three youngest deposits (in order: Cripple Creek, Oya, Kochbulak) suggest a well-defined half-life, most older samples appear to have lost  $^{130}\text{Xe}^*$  more recently than the most recent disturbance in the accepted geological record, apart from Boliden where the U-He and K-Ar systems appear to have been reset more recently than the Te-Xe system.

In Fig. 4 we compare our data with literature data. The half-life derived from our Kochbulak data is close to that derived from other young samples. Cripple Creek, with a deposit age of  $28 \pm 2$  Myr, yields a half-life of  $(9.1 \pm 0.9) \times 10^{20}$  yr [10] and Oya, with a deposit age of  $93 \pm 11$  Myr, yields a half-life of  $(8.0 \pm 1.0) \times 10^{20}$  yr [13]. Combined with our Kochbulak data we adopt a preferred value for the half-life of  $(8.0 \pm 1.1) \times 10^{20}$  yr, towards the high end of the range determined by calorimetry. Assuming the  $^{128}\text{Te}$  half-life is  $(3.52 \pm 0.11) \times 10^{-4}$  of that of  $^{130}\text{Te}$  [4] yields a half-life for  $^{128}\text{Te}$  of  $(2.3 \pm 0.3) \times 10^{24}$  yr.

The analyses of small samples that we report here yield data that are, where similar samples have been analyzed, consistent with literature data obtained from samples  $\sim 100$  times more massive. Our  $^{130}\text{Xe}^*/^{130}\text{Te}$  ratio for Boliden tellurobismuthite is an exception in being 30% lower than the literature data, perhaps reflecting some heterogeneity in this deposit. For relatively young samples ( $<300$  Myr) the evidence suggests that telluride minerals are capable of quantitative retention of xenon produced by *in situ* decay of  $^{130}\text{Te}$ . Samples from three different deposits in this age range, and samples of three different minerals from the same deposit, are consistent with retention of xenon since formation and a well defined half-life. Older samples consistently fail to reflect ancient formation ages in their Te-Xe systematics and show evidence of redistribution and loss of  $^{130}\text{Xe}^*$  in evolved compositions of associated trapped fluids. We suggest that this is not surprising; the extended geological history of samples formed in the Precambrian is significantly less well constrained than that of more recent samples. The susceptibility of older samples to

partial or complete resetting, such as major regional tectonic events, revealed by the data suggests that there is little hope of evaluating variations in the solar neutrino flux through analysis of ancient telluride minerals.

There is more hope for the dating technique. Determination of the time of precipitation of a mineral deposit allows the fluid source (and the geological activity of which it formed a part) to be constrained, allowing mineral exploration strategies to also be better constrained. However, the age of many mineral deposits remains uncertain because the minerals that make up many deposits (rather than those in the surrounding rock) are not amenable to dating by conventional radioisotope chronometers. Using the Te-Xe technique we can, for instance, confidently rule out the hypothesis that telluride deposition at Good Hope was associated with volcanic activity in the last 30 Myr [18]. However, as for any isotopic system, establishing

its authenticity and the validity of the derived ages requires analyses of multiple samples from the same deposit along with a detailed understanding of the sequence of events that have occurred in the history of the region to determine primary ages and to investigate the effects of post-formational processes.

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