Purification of Telluric Acid for SNO+ Neutrinoless Double-beta Decay Search

S. Hans^a, R. Rosero^a, L. Hu^a, O. Chkvorets^b, W.T. Chan^a, S. Guan^a, W. Beriguete^a, A. Wright^d, R. Ford^c, M.C. Chen^d, S. Biller^e, and M. Yeh^{a*}

^a Brookhaven National Laboratory, Upton, New York 11973⁺

^b Laurentian University, Sudbury, Canada

^c SNOLAB, Creighton Mine, Sudbury, Canada

^d Queen University, Kingston, Canada

^e University of Oxford, Oxford OX1 3RH, UK

Abstract. Tellurium-130 has the highest natural abundance of any double-beta decay isotopes. Recently it has been developed as a promising candidate for loading in liquid scintillator to explore the Majorana or Dirac nature of the neutrino through a search for neutrinoless double beta decay ($0\nu\beta\beta$). To this end, procedures have been developed to transfer tellurium ions into the organic liquid by a water-based loading technology. However, traces of naturally occurring radioactivity and cosmic-ray induced isotopes introduced into the scintillator with tellurium could produce undesirable contaminations in the ^{130}Te $0\nu\beta\beta$ region. Measurements using various elemental spikes prepared from different chemical forms indicate that the uses of self-scavenging as well as acid and thermal recrystallization prior to the preparation of a tellurium-loaded liquid scintillator can deplete U and Th and several cosmic-activated isotopes from Te feedstock by a factor of $10^2 - 10^3$ in a single pass. The process is also found to improve the optical transmission in the blue region, sensible to the photomultiplier tube, by removing traces of colored impurities. In addition to the scintillator-based experiments, this cleansing scheme has potential applications to the production of radiopure tellurium crystals for other rare-event experiments.

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* Corresponding author. +01-631-344-2870; fax: +01-631-344-5815; E-mail address: yeh@bnl.gov (M. Yeh)

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1. INTRODUCTION

Much has been learned about neutrino mixing mechanism over recent years¹⁻³, yet there are still a number of neutrino properties that remain unknown. These include the absolute magnitude of neutrino mass, the extent of CP violation, and the Majorana or Dirac nature of neutrinos. Attempts to measure the mass scale and test for the Majorana neutrino via a rare radioactive decay process known as neutrinoless double beta decay ($0\nu\beta\beta$) have spanned decades from the early methods of radiochemical extraction to the recent technologies of semiconductor, bolometer, real-time scintillator (crystal, liquid, or sandwich-foil), or tracking TPC⁴. These various detection schemes can be separated by their approaches to distinguishing differences in the summed electron energy spectrum near the decay endpoint, where backgrounds from internal and external radioactivity need to be greatly suppressed. Among a variety of the proposed experiments, SNO+ and KamLAND-Zen (KZ) are the only scintillator-based detectors in which a $0\nu\beta\beta$ candidate is loaded in scintillation liquid to serve as detection medium and source⁵⁻⁶.

In principal, any double beta decay (DBD) isotopes soluble in the liquid scintillator can serve as the source for $0\nu\beta\beta$ searches. Although the energy resolution of such a scintillator detector will not be as good as competing solid-state or bolometric detectors, the large self-shielding volumes of high purity liquid and potential to load significant amounts of isotope could more than compensate for this deficiency. Furthermore, liquid scintillator detectors are readily scalable and have significant flexibility in the configuration in terms of loading levels and alternative deployments of $0\nu\beta\beta$ isotope as well as in-line purifications.

Yet for some $0\nu\beta\beta$ isotopes (e.g. calcium, tellurium, selenium, molybdenum, and germanium), it is not feasible to employ the conventional complexing-ligand, -COOH or -P=O, organometallic loading method for a large production⁷ due to their hydrophilic natures. Recently, a water-based loading technology has been developed to offer an alternate approach of loading such isotopes in an organic scintillator⁸⁻⁹. This permits the use of organometallic liquid-scintillator in particle physics experiments and has offered an opportunity for SNO+ to deploy ¹³⁰Te as a $0\nu\beta\beta$ isotope in its detector¹⁰⁻¹¹.

Tellurium-130 is of great interest as a $0\nu\beta\beta$ source owing to its low $2\nu\beta\beta$ rate, good optical transparency in the visible region, and high natural abundance (34.1%, $Q=2529$ -keV), which negates the need for costly enrichment. In addition, tellurium compounds, as an element in the Group-VIA family, typically have relatively low actinide contaminations compared with lanthanide isotopes (e.g. Nd). However, even though Te has low intrinsic U/Th backgrounds, the cosmogenic production of radioactive isotopes, such as sodium (²²Na), cobalt (⁶⁰Co), silver (^{110m}Ag), antimony (^{124,126m}Sb), and tin (¹²⁶Sn), could still result in backgrounds to the $0\nu\beta\beta$ region¹². Some of these cosmogenic isotopes, i.e. Sn, are not soluble in water due to their high hydrolysis constants and thus can be removed along with U and Th by the scavenging method (3- μ m filtration at pH>4) that was previously developed for gadolinium (Gd), neodymium (Nd), and applicable to other lanthanides via their feeding solutions¹³. However Co, Ag, and many other cosmogenic isotopes are very water soluble, which implies so this scavenging method is not effective for their removal. Thus, care must be taken to cleanse these

cosmogenic and radiogenic contaminants from the starting materials so as not to introduce them during the metal-loading process.

This paper describes a practical purification process consisting of self-scavenging plus a combination of both acid and thermal induced recrystallization to remove radioactive contaminants from telluric acid, $\text{Te}(\text{OH})_6$. Unlike the use of TeO_2 crystals by the bolometric cryogenic experiments (e.g. CUORE), telluric acid, a refined product from tellurium oxide or chloride, is selected as the starting material for the SNO+ $0\nu\beta\beta$ experiment due to its (1) lower intrinsic radioactive impurities, compared to TeO_2 , through the additional refinement process and (2) high solubility in water, making it easier to purify further and incorporate into the LS loading procedures either by conventional complexing-ligand method or by the new water-based technology. Note that a simple oxidation reaction from either hydrogen peroxide addition or gentle heating to evaporate the water of the purified tellurium-acid aqueous solution would convert the $\text{Te}(\text{OH})_6$ to TeO_2 , which could then be used for the production of radiopure tellurium crystals for other $0\nu\beta\beta$ experiments if desired.

2. EXPERIMENTAL PROCEDURES

Crystals of telluric acid, $\text{Te}(\text{OH})_6$, from ABSCO and Sigma-Aldrich were used in the experiments. Nitric acid (HNO_3 , ACS-grade 70%) and hydrochloric acid (HCl , ACS-grade 37%) were purchased from Sigma. Ammonium hydroxide (NH_4OH , ACS-grade 20-22% as NH_3) was obtained from BDH. Ultrapure 18-M Ω .cm water (UPW) was acquired from a Millipore Milli-Q system. Pilot OPTIPHASE-HiSafe3 was used as the scintillation cocktail. Two types of measurements are described in the sub-sections below: (1) a chemical analysis of Co, Sb, Sn, Y, Sc, Zr, and Ag traces spiked in $\text{Te}(\text{OH})_6$ before and after purifications by, and (2) a radiotracer study of Th and its daughters removed from, scavenging and acid/thermal recrystallization. An Oxford β - α coincidence counter¹⁴ situated at Laurentian University (Sudbury, Canada) was used for radiotracer analysis, while all other non-radioactive spike analysis, e.g. x-ray Fluorescence (XRF), Inductively Coupled Plasma Mass Spectrometry (ICP-MS), and UV-Vis spectrometry, were performed at Brookhaven National Laboratory (Upton, USA).

2.1. Scavenging and Acid-Recrystallization

Preparation of each spiked solution was completed in a 15% tellurium (w/w) aqueous solution by dissolving telluric acid in water with the addition of concentrated hydrochloric acid (12N) to ensure full dissolution of the telluric crystals and complete dissociation of spiked elements from their chemical forms. As the telluric acid was first introduced into the water, the solution was slightly cloudy with few fine particles floating in the solution. Upon the addition of a few drops of HCl , the solution turned clear. Each spiked element of Co, Sb, Sn, Sc, Y, Zr, or Ag, prepared from different chemical compounds in chloride, nitrate, oxide, or sulfide in UPW, was added to this 15%Te- H_2O solution separately. A few grams of the spiked 15% tellurium solution (SS) were taken out for analysis as the baseline for comparison. The rest of the spiked tellurium solution was scavenged first by pH-adjustment, followed by filtration (3- μm pore; Whatman-44).

Approximately 5 grams of the scavenged filtrate were again taken aside for analysis (SF). Lastly 1:1 ratio of concentrated nitric acid (15N) was added to the excess tellurium spiked filtrate to (1) ensure a complete dissolution of all elements regardless of initial chemical forms and (2) induce the recrystallization of the telluric acid in high purity. Note that this designated method is unique and works only for the telluric acid compounds.

The resulting HNO₃-added solution (RM) was allowed to settle at room temperature for an hour. Within few minutes, a white solid of telluric acid appeared in the solution. Vacuum filtration was used to separate out the recrystallized telluric acid from the residual spiked liquid (RS). The solid was washed 3 times with ethanol to remove any traced spikes. The washed liquid (RW) was also collected for mass-balance check.

2.2. The ²²⁸Th Radiotracer

An Eckert-Ziegler solution of ²²⁸Th, t_{1/2}=1.913 years, was used to simulate the behavior of natural ²³²Th in the Te(OH)₆ system. The ²²⁸Th tracer was certified to have the activity of 3707 Bq/mL in equilibrium with its daughters in April 2009. The 6α-3β decay chain leading from ²²⁸Th to stable ²⁰⁸Pb is shown in Figure 1 (5α-2β in account for the decay branches as ²¹²Bi). Half-lives and some α-, β- energies of interest are included in the figure. The chain members, ²²⁸Th, ²²⁴Ra, ²¹²Bi and ²¹²Pb enclosed in boxes in Figure 1, serve as tracers for their respective elements. Ten μL of the ²²⁸Th tracer (10.73 Bq) were first spiked into a 15% Te-H₂O solution. Then, 0.5 mL of the Th-spiked Te solution (TeA0) was taken for counting by the Oxford β-α coincidence counter. After acid-recrystallization, the purified Te(OH)₆ solid was dissolved in UPW and counted (sample-TeA1). The filter used for recrystallization was also cut in small pieces of ~3 mm for counting (sample-TeA2). All samples were counted for 220 hours.

3. RESULTS AND DISCUSSION

3.1. Non-tracer Chemical Analysis

3.1.1. Cobalt (⁶⁰Co) and Authentication of X-ray Fluorescence Spectrometry

X-ray fluorescence spectrometry (XRF) measures the emission of characteristic x-rays (secondary) from an element that has been excited by higher energies of either x- or γ-ray. This non-destructive XRF scan could simplify the sample preparation for measurements and was used to measure the concentration of metal ion for this study. Its sensitivity and verification of instrumental norms are described below.

Cobalt-60 is one of the main background threats, due to its γ-sum energies, which could overlap with the Q value of ¹³⁰Te 0νββ decay. The cobalt is very water-soluble and cannot therefore be removed effectively by scavenging. The ~1% Co spike in Te-H₂O was measured to be 9915±343 ppm (SL). The filtrate (MF) after recrystallization gave 8540±188 ppm of Co in water; while the trace in the ethanol-washed liquid (MW) was 955±11ppm. The cobalt trace lost in the filter paper was known to be 5% by monitoring

the trace loss of repeating filtrations in several trails continuously. The Co content in the purified $\text{Te}(\text{OH})_6$ solid (MP) after recrystallization was measured to be 6.10 ± 2.16 ppm. The sum of Co content in MF, MW and MP, plus the 5% loss from filtration balances the total Co trace (SL) spiked into the initial 15% $\text{Te-H}_2\text{O}$. The reduction factor of Co removed by the $\text{Te}(\text{OH})_6$ recrystallization, a ratio of SL/MP, was then calculated to be $(1.62 \pm 0.58) \times 10^3$. Similar studies were repeated six times and an average reduction factor of $(1.15 \pm 0.66) \times 10^3$ was obtained. For cross-check, the MP was sent for ICP-MS analysis [Robertson Microlit Laboratories, NJ] and no trace of Co was detected, resulting in a limit of <10 ppm from this analysis, which is in agreement with the XRF measurement.

To further understand and cross-check the sensitivity of XRF, a standard-addition method was applied to verify the results from the above direct-counting method of cobalt concentration. A series of known Co-standards from 1 to 20 ppm was added to the MF separately, and each sample was counted by XRF to a statistical precision of $\pm 1\%$. The intensity of emission fluorescence is a linear function of concentration in the region of up to 100 ppm; the Y-intercept is interpreted as the original cobalt content in the MF. Figure 2(a) shows the measured concentration of Co (in ppm; converted from fluorescence counts internally by instrument) at each standard-added cobalt solution. The emission spectra of Co-spikes in Te before and after recrystallization are presented in Figure 2(b). The calculated Y-intercept of 9.49 ppm is consistent to within 2% in comparison with the direct-counting of 9.64 ppm of MF (corresponding to 8540 ppm after dilution-factor correction). The results indicated that metal-concentration determined by XRF is consistent from two independent approaches. We then use the direct counting method to quantify the reduction factors for all elements described in the following sub-sections.

3.1.2. Tin (^{126}Sn), Zirconium (^{88}Zr)

Tin and zirconium have the pK_{sp} values of 10^{-56} and 10^{-49} respectively at room temperature indicating their very low solubility in water, thus that both elements can be depleted from Te compound by the pH-scavenging method¹⁵. The 15% $\text{Te}(\text{OH})_6$ dissolved in UPW gave a pH of ~ 2.71 . After adding the spikes of either Sn (0.1%) or Zr (0.1%) into this 15% $\text{Te-H}_2\text{O}$, a thin-layer of precipitate was formed immediately. The precipitates were removed by a $3\text{-}\mu\text{m}$ filter, and the residual filtrate had no detectable traces of Sn or Zr by XRF (see Figures. 3(a) & (b)). Based on the XRF detection limits (σ)¹⁶ of 6ppm for Sn and 3.6ppm for Zr, the reduction factor was determined to be $>1.67 \times 10^2$ for Sn and $>2.78 \times 10^2$ for Zr.

3.1.3. Silver ($^{110\text{m}}\text{Ag}$), Yttrium (^{88}Y), Scandium (^{46}Sc), Antimony ($^{124,126\text{m}}\text{Sb}$)

Silver, yttrium, scandium and antimony are other potential contributors of background to the $0\nu\beta\beta$ measurements due to their long half-lives and large electron energies close to the Q-values. Among them, $^{110\text{m}}\text{Ag}$ ($t_{1/2}=249.8\text{d}$) has been identified by CUORE's measurements of production cross sections by proton and neutron beams at LANSCE¹⁷⁻¹⁸ and is also observed as a $0\nu\beta\beta$ background with few possible sources of contamination by the KamLAND-Zen experiment⁶. These four potential cosmogenic impurities have modest solubility in water; thus only negligible traces can be removed by scavenging. Consequently, approximately 0.1% of Ag ($\sigma \sim 3.6\text{ppm}$) and/or Y ($\sigma \sim 3.6\text{ppm}$)

was spiked in the 15% Te-H₂O solution, respectively, to investigate their removal efficiencies by acid recrystallization. On the other hand, Sc ($\sigma \sim 18$ ppm) and Sb ($\sigma \sim 12$ ppm) have lower sensitivities by XRF counting; therefore higher spikes of 0.3% for Sc and/or Sb were used. The XRF fluorescence spectra of Ag, Y, Sc, and Sb before and after recrystallization are shown in Figures 4(a)-(d). No trace of each spiked element was detected after purification from the XRF emission scan. The reduction factor based on each sensitivity limit was calculated as $>2.78 \times 10^2$ for Ag, $>2.78 \times 10^2$ for Y, $>1.65 \times 10^2$ for Sc and $>2.43 \times 10^2$ for Sb based on individual sensitivity.

An ancillary benefit of scavenging and acid-recrystallization is the improvement of optical property. Yellowing of a metal-loaded scintillator by traces of colored impurities could deteriorate the optical transparency by reducing blue light transmission or affect the chemical stability. Figure 5 shows that the optical transparency of the Te(OH)₆ solution has been significantly improved after purification.

3.2. Tracer Studies

Decay rates of the ²³²Th-chain daughters from ²²⁸Th to ²⁰⁸Pb are equal in an equilibrated system. However the chemical fractionation during the partial hydrolysis and recrystallization can break this equilibrium, so measurements of various β - α coincidences are needed to quantify each fractionation. The thorium scavenging has been previously studied using Gd- and Nd- loaded liquid scintillator from pH 4 - 6¹³. Since the starting pH of Te(OH)₆ in water is below 4, no scavenging test for Th removal from the Te-H₂O has been done. In this study, we focused only on the feasibility of removing Th and its daughters from Te(OH)₆ by acid-recrystallization. The introduced activity was calculated to be 10.73Bq of ²²⁸Th per 4.25g of telluric acid, or 2.52 Bq/g. The measured initial activities of the sample (TeA0) were 2.61 ± 0.03 Bq/g for ²²⁸Th, 2.70 ± 0.03 Bq/g for ²²⁴Ra, 2.66 ± 0.03 Bq/g for ²¹²Pb, and 2.68 ± 0.03 Bq/g for ²¹²Bi, as expected in an equilibrium system. The systematic error was due to the uncertainty in the counting efficiency, which was of the order of $\sim 10\%$. After recrystallization, the activities of purified telluric acid (TeA1) were 6.7 ± 0.3 mBq/g for ²²⁸Th, 6.8 ± 0.3 mBq/g for ²²⁴Ra, 8.9 ± 0.7 mBq/g for ²¹²Pb and 7.7 ± 1.8 mBq/g for ²¹²Bi. The Th and Ra were in equilibrium after recrystallization, while the excess of Pb and Bi could be from either the migration of radon (²²⁰Rn) during the preparation, or different fractional distributions for different elements using the recrystallization method. The reduction factors were calculated as 390 ± 19 for ²²⁸Th, 397 ± 20 for ²²⁴Ra, 299 ± 22 for ²¹²Pb and 348 ± 81 for ²¹²Bi. The recovery of Te(OH)₆ solid after recrystallization was found to be better than 95%, which is in agreement with that of the chemical analysis.

A summary of reduction factor obtained from trace and non-trace analysis is presented in Table 2.

3.3. Double-Pass Acid Recrystallization

The data from the acid-recrystallization of telluric acid indicate that this purification method is applicable to the removal of a wide range of elements, yielding similar reduction factors of 10^2 to 10^3 (dependent on the measurement sensitivity) in a

single pass. The potential impact of cosmogenic isotopes has been evaluated for SNO+ and suggests that a reduction factor in the range of 10^4 to 10^5 might be necessary. The possibility to achieve this using two purification passes was investigated. Cobalt was chosen as a representative element for the double-pass test due to its well-defined calibration curve and as noted above that all elements appear to have similar reduction efficiencies by recrystallization.

The ~1% cobalt-spike in 15% Te-H₂O was prepared in a 2-liter beaker (1001 g). The filtrate after recrystallization serves as an indication of how much Co content was (1) being removed from or (2) present in the Te-acid solids before and after purification. The filtrate was then inferred to the [Co] content in the Te(OH)₆ solids to avoid the interference from large Te fluorescence counts overlapping with Co signal (if the Te-solids were measured directly). In that sequence, the amount of Co measured from 2nd filtrate represents the Co associated with the Te(OH)₆ solids after first pass of purification and similarly, Co in 3rd filtrate, signifies its quantity in Te(OH)₆ solids after 2nd pass of purification. The errors by this method are from (1) traces of Co in solid-washing liquid or on filter paper (after filtration), known to be 5%; and (2) residual trace in the solids after recrystallization, estimated to be <10% from previous study of 10^3 reduction. In this study, the Te(OH)₆ solids were recrystallized three times at 1-liter scale each. The 3rd 1-liter filtrate was gradually evaporated to ~5g to enhance the measurement sensitivity by a factor of ~200.

The initial 1% Co in 15% Te(OH)₆ (270g solid) was used for the first pass. The 2nd pass used 204g of recrystallized Te(OH)₆ from first pass after purification. Lastly, 100g of Te(OH)₆ from 2nd pass was recrystallized again to obtain the filtrate of Co water (equivalent to Co in 2nd Te(OH)₆ solid). The sample vials from different passes are shown in Figure 6, where the dark purple Co spike was removed after double-pass purification. The same procedures were repeated six times, and the average reduction factor (k_n) at each pass is presented in Table 3. The Co reduction factors between first (k_1) and second pass (k_2) of Te recrystallization are consistent within the uncertainties. This study demonstrates that the double-pass acid-recrystallization followed by 3× ethanol wash reduces the Co (analog to other cosmogonies besides Te) by a factor of 10^6 .

3.4. Thermal Recrystallization

The acid-recrystallization combined with a final rinse of ethanol effectively removes the cosmogenic and radiogenic isotopes and cleanses the residual traces from the telluric acid crystals. However, practically such purification sequences can likely only be applied at a surface facility due to the potential fire hazard of ethanol in an underground space. After purification on surface and before transportation underground, there will potentially be a few hours of cosmic exposure on the double-pass recrystallized telluric acid, which would regenerate diminutive cosmogenic background from few short half-life isotopes, such as Ag, Co, Sb, Y or Zr. Since these cosmic-regenerated isotopes from short-term exposure are in very small quantities, an additional factor of 10^2 reduction is required once transferred into underground.

336 Considering the hazardous chemicals are difficult to use underground, a different
337 approach to the purification by acid-recrystallization using ultrapure water and thermal
338 control is proposed. The purified telluric acid, after transferring from surface to
339 underground, will be first dissolved in water at 80°C in a thermal bath for an hour to
340 ensure a complete dissolution of all refined telluric crystals. Taking advantage of the fact
341 that solubility is a function of temperature, the telluric aqueous solution will be then
342 filtered and transferred to an ice bath for cold re-precipitation.

343
344 To test this new procedure, individual 0.05% spikes of Ag, Co and/or Zr was
345 added to 15% Te aqueous solution during the crystal dissolution in the heated thermal
346 bath, and the purification of scavenging (pH=2.8, 3µm filtration) followed by thermal
347 recrystallization was applied after full mixing. The Zr was removed effectively by
348 scavenging (see Figure 7), corresponding to a reduction limit of $>1.44 \times 10^2$ based on the
349 XRF sensitivity ($\sigma \sim 3.6$ ppm). The Ag has no detectable trace after thermal
350 recrystallization; thus a reduction limit of $>1.44 \times 10^2$ ($\sigma \sim 3.6$ ppm) is given. The Co
351 reduction factor after thermal recrystallization was found to be $(1.04 \pm 0.85) \times 10^2$, with the
352 large deviation due to the quantity variations of Te water after 80°C heating and the
353 temperature of ice bath for cooling. Table 4 summarizes the reduction factors for Ag, Co
354 and Zr. Approximately 70% of telluric acid crystal was regenerated from thermal
355 recrystallization. For SNO+, the residual 30% of telluric acid solution will be transferred
356 back to the surface for repeated acid recrystallization, and the cycle will be continuing
357 until all crystals are purified.

358 359 4. CONCLUSIONS 360

361 We have shown that scavenging and single-pass acid-recrystallization of $\text{Te}(\text{OH})_6$
362 reduce the radiogenic and cosmogenic isotopes by a factor of 10^2 to 10^3 with a few %
363 loss of tellurium. Undesirable colored impurities are also reduced via these purification
364 methods. The required factor of reduction for SNO+ depends on the purity of starting
365 tellurium compounds. A recent radioisotopic measurement of TeO_2 from the CUORE
366 experiment shows only appreciable level of Sb and Ag found in the tellurium crystal¹⁹.
367 The exposure was too short to have measurable levels of Co. However, other proton
368 activation experiments show that Co can be produced in TeO_2 ¹⁶. SNO+ takes the
369 precaution of identifying and attempting to remove as many potential radiogenic and
370 cosmogenic isotopes that could contribute to the ^{130}Te $0\nu\beta\beta$ region as possible. The
371 presented data demonstrate that Co (analog for other impurities) can be effectively
372 removed by a factor of 10^3 using the proposed single-pass, and 10^6 by double-pass
373 purification. The operation of this self-scavenging followed by acid-recrystallization is
374 simple, cost-effective, and easily adapted for the large-scale production of liquid
375 scintillator loaded with tellurium (Figure 8). The whole purification cycle can be
376 completed within hours, such that, purified telluric acid is ready for either production of
377 metal-loaded liquid scintillator or storage at underground to allow further decay of any
378 radioisotopes with short half-lives before synthesis. To remove the cosmic-regenerated
379 isotopes from the refined $\text{Te}(\text{OH})_6$ after few hours of exposure above ground, a thermal
380 recrystallization procedure using pure water has also been developed. A reduction factor
381 of 10^2 or better is observed for Ag, Co, and Zr.
382

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Table 1: Background isotopes that can be produced by spallation reactions on ^{nat}Te target induced by cosmic neutrons and protons at sea-level (adapted from ref. 12).

Isotope	$T_{1/2}$ [d]	Q-value [MeV]	Events in 1 yr after 1-yr surface exposure
^{60}Co (direct and daughter of ^{60}Fe)	1925.27 (5.48E+8)	2.82	6.91e3
^{88}Y (direct and daughter of ^{88}Zr)	106.63 (83.4)	3.62	3.91e5
$^{110\text{m}}\text{Ag}$ (98.67% direct with 1.33% IT to ^{110}Ag)	249.83	3.01	6.96e4
^{110}Ag (daughter of $^{110\text{m}}\text{Ag}$ at 1.33% IT decay)	2.85E-4	2.89	936
^{124}Sb	60.2	2.90	3.10e6
^{44}Sc (direct and daughter of ^{44}Ti)	0.17 (2.16E+4)	3.65	57.5(direct)+44.5
^{46}Sc	83.79	2.37	4.36e4
^{68}Ga (daughter of ^{68}Ge)	4.70E-2(270.95)	2.92	3.73e4
^{26}Al	2.62E+8	4.00	0.05
^{82}Rb (daughter of ^{82}Sr)	8.75E-4(25.35)	4.40	1.80e4
^{42}K (direct and daughter of ^{42}Ar)	0.51 (1.20E+4)	3.53	199(direct)+366
^{56}Co	77.24	4.57	2.63e3
^{58}Co	70.89	2.31	2.52e4
^{106}Rh (daughter of ^{106}Ru)	3.47E-4 (371.8)	3.54	1.53e3
$^{126\text{m}}\text{Sb}$ (direct daughter of ^{126}Sn . 86% β^- decay and 14% IT to ^{126}Sb)	0.01 (8.40E7)	3.69	238.5(direct)+1.5
^{126}Sb (direct and daughter of $^{126\text{m}}\text{Sb}$ (14%))	12.35 (0.01)	3.67	3.59e5
^{22}Na	950.64	2.84	1.53e4
^{84}Rb (96.1% EC decay with high Q-value, the rest is a β^- decay with lower Q-value)	32.82	2.69	1.18e4
^{90}Y (direct and daughter of ^{90}Sr)	2.67 (1.05E+4)	2.28	537(direct)+286
^{102}Rh (78% EC decay with high Q-value and daughter of $^{102\text{m}}\text{Rh}$ at 0.23% IT decay)	207.3	2.32	2.76e5
$^{102\text{m}}\text{Rh}$ (99.77% EC decay, 0.23% IT to ^{102}Rh)	1366.77	2.46	1.34e5

Isotopes in list are either directly produced in the target or come from the decay of parent isotopes produced in the target.

Table 2: Summary of reducing factor for selected cosmogenic isotopes using single-pass acid recrystallization

Isotope	XRF Sensitivity (ppm) ³	Reducing Factor
Sn ¹	6.0	$>1.67 \times 10^2$
Zr ¹	3.6	$>2.78 \times 10^2$
Co ¹	1.5	$(1.15 \pm 0.66) \times 10^3$
Ag ¹	3.6	$>2.78 \times 10^2$
Y ¹	3.6	$>2.78 \times 10^2$
Sc ¹	18	$>1.65 \times 10^2$
Sb ¹	12	$>2.43 \times 10^2$
²²⁸ Th ²	-	$(3.90 \pm 0.19) \times 10^2$
²²⁴ Ra ²	-	$(3.97 \pm 0.20) \times 10^2$
²¹² Pb ²	-	$(2.99 \pm 0.22) \times 10^2$
²¹² Bi ²	-	$(3.48 \pm 0.81) \times 10^2$

¹X-ray fluorescence chemical analysis.

²By ²²⁸Th radiotracer.

³Thermo ARL QUANT'X database¹⁶.

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Table 3. Double Pass Average Test Results of Cobalt

	k_n
1 st Pass (k_1)	$(1.15 \pm 0.66) \times 10^3$
2 nd Pass (k_2)	$(0.93 \pm 0.89) \times 10^3$

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Table 4. Summary of Reduction Factor for Selected Elements using Scavenging and Thermal Recrystallization

	K_1
Ag	$>1.44 \times 10^2$
Co	$(1.04 \pm 0.85) \times 10^2$
Zr	$>1.44 \times 10^2$

Figure 1

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^{90}Th			<div>228 1.913y α5520.08</div>
^{89}Ac			↓
^{88}Ra			<div>224 3.657d α5788.85</div>
^{87}Fr			↓
^{86}Rn			<div>220 0.927m α6404.67</div>
^{85}At			↓
^{84}Po	<div>212 299ns α8954.12</div>		<div>216 145ms α6906.32</div>
^{83}Bi	↓	<div>212 60.6m α6207.26 β2252.13</div>	↓
^{82}Pb	<div>208 Stable</div>	↓	<div>212 10.64h β569.91</div>
^{81}Tl		<div>208 3.053m β4998.98</div>	

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Figure 2a

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Figure 2b

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Figure 3a

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Figure 3b

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Figure 4a

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Figure 4b

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Figure 4c

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Figure 4d

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Figure 5

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Figure 6

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Figure 7

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Figure 8

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Surface Facility (Acid-recrystallization)

- Dissolve $\text{Te}(\text{OH})_6$ in water
- Recrystallize using nitric acid
- Rinse with ethanol

Underground Facility (Thermal-recrystallization)

- Dissolve purified $\text{Te}(\text{OH})_6$ in 80°C water
- Recrystallize using cold water
- Filter $\text{Te}(\text{OH})_6$ refined crystal (70%) for storage; then production

Figure Captions

Figure 1: ^{228}Th decay chain, red boxes are selected isotopes for coincidence counting.

Figure 2a: Determination of Co-content in the ML by standard-addition method.

Figure 2b: Comparison of XRF spectra for Te ($L_{\alpha,\beta}$ =3.77-keV, 4.03-keV) with Co-spike ($K_{\alpha,\beta}$ =6.93-keV, 7.65-keV) before and after recrystallization.

Figure 3a: Comparison of XRF spectra for Te ($K_{\alpha,\beta}$ =27.38-keV, 31.13-keV) with Sn-spike ($K_{\alpha,\beta}$ =25.19-keV, 28.60-keV) before and after recrystallization.

Figure 3b: Comparison of XRF spectra for Te ($K_{\alpha,\beta}$ =27.38-keV, 31.13-keV) with Zr-spike ($K_{\alpha,\beta}$ =15.74-keV, 17.69-keV) with some Y presence ($K_{\alpha,\beta}$ =14.93-keV, 16.75-keV) before and after recrystallization.

Figure 4a: Comparison of XRF spectra for Te ($K_{\alpha,\beta}$ =27.38-keV, 31.13-keV) with Ag-spike ($K_{\alpha,\beta}$ =22.10-keV, 24.99-keV) before and after recrystallization.

Figure 4b: Comparison of XRF spectra for Te ($K_{\alpha,\beta}$ =27.38-keV, 31.13-keV) with Y-spike ($K_{\alpha,\beta}$ =14.93-keV, 16.75-keV) before and after recrystallization.

Figure 4c: Comparison of XRF spectra for Te ($L_{\alpha,\beta}$ =3.77-keV, 4.03-keV) with Sc-spike ($K_{\alpha,\beta}$ =4.09-keV, 4.46-keV) before and after recrystallization. The Sc X-ray overlaps with Te; thus the ratio of peak intensities of Te L_{α} and L_{β} is fixed to de-convolute the Sc peak intensity.

Figure 4d: Comparison of XRF spectra for Te ($K_{\alpha,\beta}$ =27.38-keV, 31.13-keV) with Sb-spike ($K_{\alpha,\beta}$ =26.27-keV, 29.85-keV) before and after recrystallization.

Figure 5: Optical spectra of a 15% $\text{Te}(\text{OH})_6$ solution before and after recrystallization.

Figure 6: Picture of $\text{Te-H}_2\text{O}$ with cobalt spike (1%) at different stages of purification, dark (original Co-spike), pink (after single-pass), and clear (after double-pass).

Figure 7: Comparison of XRF spectra for Te ($L_{\alpha,\beta}$ =3.77-keV, 4.03-keV) with Zr-spike (K_{α} =15.75-keV) before and after scavenging.

Figure 8: The proposed purification schemes for $\text{Te}(\text{OH})_6$ feedstock at surface and underground facilities.