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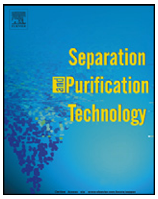

# Purification of neutron-irradiated tellurium targets from cross-radiocontaminants by precipitation with 1,10-phenanthroline

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ABSTRACT

Irradiated tellurium targets were purified by precipitation from 6 M HCl solution using 1,10-phenanthroline. The effect of molar ratio of the reactants (initial 1,10-phenanthroline:Te molar ratio) on the precipitation yield of radiotellurium was studied. It was found that, >99% of radiotellurium has been taken up by the precipitate formed with the initial 2 phenanthroline:1Te molar ratio along with different decontamination ratios from the other cross-radiocontaminants; ≥99.9% of $^{65}$Zn, 97.7 ± 1.2% of $^{75}$Se, ≥99.9% $^{110m}$Ag, and 95.4 ± 2.5% of $^{124}$Sb. Traces of $^{51}$Cr, $^{54}$Mn, $^{59}$Fe, and $^{60}$Co have been detected only in the supernatant after the precipitation process. Un-irradiated tellurium was precipitated with 1,10-phenanthroline under the same conditions with the initial 2phenanthroline:1Te molar ratio and, then, the precipitate formed was characterized by IR spectroscopy and thermal analysis. The most probable chemical composition of this precipitate was $[Te(phen)_2Cl_4]\cdot 3.4H_2O$.

## 1. Introduction

Tellurium targets are used for production of (i) $^{131}$I via $^{130}Te(n,\gamma)^{131}I$ nuclear reactions by irradiation in nuclear reactors [1] and (ii) $^{121}$I, $^{123}$I, and $^{124}$I via $^{122}Te(d,3n)^{121}I$, $^{122}Te(d,n)^{123}I$, $^{124}Te(p,2n)^{123}I$, and $^{124}Te(p,n)^{124}I$ nuclear reactions, respectively, by irradiation in cyclotrons [2–5]. Purification and recovery of the irradiated tellurium targets, after separation of radioiodine isotopes, for recycling [6] may be effective in (i) saving costs, specially in the case of isotopic-enriched Te targets, (ii) waste minimization, and (iii) possible use of the separated radiocontaminants as byproducts in different applications, e.g., preparation of radioactive mixed sealed sources (or single-sealed sources after separation from each other) used for calibration purposes. The common trace impurities, even in high-purity tellurium, include Na, Mg, Ca, B, Al, In, Si, Sn, Pb, As, Sb, Bi, Se, Ti, V, Cr, Mn, Fe, Co, Ni, Pd, Cu, Ag, Zn, and Cd [7,8]. Table 1 compiles important (n,γ) nuclear reactions of some of these impurities, discarding those producing radionuclides with half-lives of less than 1 d.

Kudelin et al. [6] reported a procedure for purification of isotopically enriched $TeO_2$, for re-use in $^{123}$I production, in which contaminated $TeO_2$ was evaporated under vacuum and condensed on a cooling finger. The $TeO_2$ was recovered with 95–97% yield and highly reduced levels of impurities of Al, Fe, Mg, Ca, Cu, Ti, Zn, Mn, Cr, B, Sn, Mo, Sb, V, Nb, Zr, La, and Be.

Lenher and Smith [14] described what is called Scott's distillation method for separation of selenium from tellurium, which was based on the volatility of selenium chloride from sulfuric acid solution when treated with HCl gas while tellurium chloride is nonvolatile under the same conditions.

Betti et al. [15] investigated separation of tellurium and selenium using divinylbenzene–styrene sulfonated copolymer cation-exchange columns, latex agglomerated anion-exchange columns, as well as multiphase cation- and anion-exchange/reversed columns and utilizing 2–220 mM HCl as eluent.

Dixit and Khopkar [16] used a reversed-phase extraction process to separate Te(IV) from different mixtures of elements using a chromatographic column of silica gel coated with tributylphosphate, for example, from a mixture of Se(IV), Te(IV), Ga, Cr(VI) and Tl(III) (by extraction from 2.5 M HCl and stripping with 1.5 M HCl), and from a mixture of Pb, Sb(III), Te(IV), Fe(III), and Au (by extraction from 6 M HCl and stripping with 1.5 M HCl, after stripping Sb with 2.5 M HCl).

Tang et al. [17] investigated the chromatographic separation of Te and Se with two different reversed-phase chromatographic methods using chromatographic columns of P-350 (methyl-bi-isooctyl phosphate) and TBP (tributyl phosphate) as stationary phases and elution with, e.g., 1.0 M HCl + 2.5 M NaCl and 1.6 M HCl, respectively.

Mhaske and Dhadke [18] suggested separation of selenium and tellurium by liquid–liquid extraction method in which Se(IV) and Te(IV) mixture in 3.5 M HCl was equilibrated with cyanex 925 (in toluene). Se(IV) remained in the aqueous phase with a recovery

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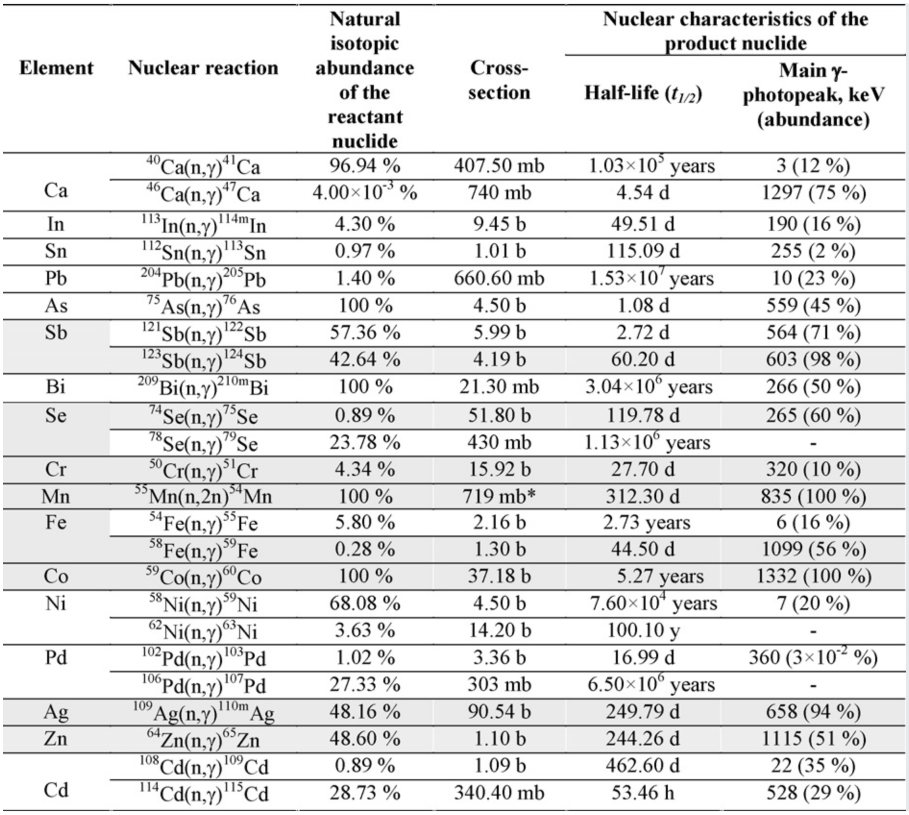
**Table 1**
Important (n,γ) nuclear reactions of some possible impurities present in tellurium targets [9–13]



| Element | Nuclear reaction | Natural isotopic abundance of the reactant nuclide | Cross-section | Nuclear characteristics of the product nuclide | |
|---|---|---|---|---|---|
| | | | | Half-life ($t_{1/2}$) | Main γ-photopeak, keV (abundance) |
| Ca | $^{40}Ca(n,\gamma)^{41}Ca$ | 96.94 % | 407.50 mb | $1.03\times10^{5}$ years | 3 (12 %) |
| | $^{46}Ca(n,\gamma)^{47}Ca$ | $4.00\times10^{-3}$ % | 740 mb | 4.54 d | 1297 (75 %) |
| In | $^{113}In(n,\gamma)^{114m}In$ | 4.30 % | 9.45 b | 49.51 d | 190 (16 %) |
| Sn | $^{112}Sn(n,\gamma)^{113}Sn$ | 0.97 % | 1.01 b | 115.09 d | 255 (2 %) |
| Pb | $^{204}Pb(n,\gamma)^{205}Pb$ | 1.40 % | 660.60 mb | $1.53\times10^{7}$ years | 10 (23 %) |
| As | $^{75}As(n,\gamma)^{76}As$ | 100 % | 4.50 b | 1.08 d | 559 (45 %) |
| Sb | $^{121}Sb(n,\gamma)^{122}Sb$ | 57.36 % | 5.99 b | 2.72 d | 564 (71 %) |
| | $^{123}Sb(n,\gamma)^{124}Sb$ | 42.64 % | 4.19 b | 60.20 d | 603 (98 %) |
| Bi | $^{209}Bi(n,\gamma)^{210m}Bi$ | 100 % | 21.30 mb | $3.04\times10^{6}$ years | 266 (50 %) |
| Se | $^{74}Se(n,\gamma)^{75}Se$ | 0.89 % | 51.80 b | 119.78 d | 265 (60 %) |
| | $^{78}Se(n,\gamma)^{79}Se$ | 23.78 % | 430 mb | $1.13\times10^{6}$ years | - |
| Cr | $^{50}Cr(n,\gamma)^{51}Cr$ | 4.34 % | 15.92 b | 27.70 d | 320 (10 %) |
| Mn | $^{55}Mn(n,2n)^{54}Mn$ | 100 % | 719 mb* | 312.30 d | 835 (100 %) |
| Fe | $^{54}Fe(n,\gamma)^{55}Fe$ | 5.80 % | 2.16 b | 2.73 years | 6 (16 %) |
| | $^{58}Fe(n,\gamma)^{59}Fe$ | 0.28 % | 1.30 b | 44.50 d | 1099 (56 %) |
| Co | $^{59}Co(n,\gamma)^{60}Co$ | 100 % | 37.18 b | 5.27 years | 1332 (100 %) |
| Ni | $^{58}Ni(n,\gamma)^{59}Ni$ | 68.08 % | 4.50 b | $7.60\times10^{4}$ years | 7 (20 %) |
| | $^{62}Ni(n,\gamma)^{63}Ni$ | 3.63 % | 14.20 b | 100.10 y | - |
| Pd | $^{102}Pd(n,\gamma)^{103}Pd$ | 1.02 % | 3.36 b | 16.99 d | 360 ($3\times10^{-2}$ %) |
| | $^{106}Pd(n,\gamma)^{107}Pd$ | 27.33 % | 303 mb | $6.50\times10^{6}$ years | - |
| Ag | $^{109}Ag(n,\gamma)^{110m}Ag$ | 48.16 % | 90.54 b | 249.79 d | 658 (94 %) |
| Zn | $^{64}Zn(n,\gamma)^{65}Zn$ | 48.60 % | 1.10 b | 244.26 d | 1115 (51 %) |
| Cd | $^{108}Cd(n,\gamma)^{109}Cd$ | 0.89 % | 1.09 b | 462.60 d | 22 (35 %) |
| | $^{114}Cd(n,\gamma)^{115}Cd$ | 28.73 % | 340.40 mb | 53.46 h | 528 (29 %) |

*Note*: cross-section of the nuclear reaction at 14 MeV neutrons, otherwise at 0.0253 eV. Highlighted rows include the radiocontaminants appeared in the γ-ray spectra of the irradiated tellurium solutions in this work.

yield of 99.1%, while Te(IV) was extracted in the organic phase and then stripped with 6 M $HNO_3$ with a recovery yield of 99.4%.

Downs and Miller [19] reported a procedure for separation of antimony and tellurium, in which $^{118}$Te was separated from the proton-irradiated antimony by dissolving the irradiated Sb target in 9 M HCl, dilution to 2 M HCl, addition of 3.5 mM $OCl^-$, and loading onto Dowex 1-x8 anion-exchange column. The column was washed with further 5 ml of 2 M HCl and then radiotellurium was eluted with deionized water.

Rai and Kukreja [20] stated a thin-layer chromatographic separation method for Au, Se, Te, Pt, and Pd by mixing their mixture solution with 2% diethyldithiocarbamate, raising pH to 8, and extracting the precipitate formed with 10 ml of chloroform. After application to a silica gel coated plate, development with benzene, toluene, or xylene eluent achieved a good separation of these elements.

Tellurium and selenium could be separated from iron by reduction in an acid solution with $SnCl_2$. Tellurium could be separated from selenium by adding bromine to HCl solution of the elements and precipitating selenium with $SO_2$. In addition, information was given that $Te^{6+}$ could be quantitatively carried on $Fe(OH)_3$ in a strong ammonia solution and thus effectively separated from $Se^{6+}$ which is not coprecipitated with $Fe(OH)_3$ under these conditions [21].

Rickard and Wyatt [22] reported the separation of tellurium from selenium and other fission products from HCl and HBr solutions by using 1,10-phenanthroline as a precipitating agent after addition of tellurium carrier, but without further details neither about the recovery yield and decontamination ratios from other nuclides in the reaction media nor about concentration of 1,10-phenanthroline and tellurium carrier. In addition, no information was mentioned about the chemical species of the separated nuclides left in the supernatant after the precipitation process.

This work aims mainly at (i) purification of the neutron-irradiated tellurium target (used for $^{131}$I production) from the cross-radiocontaminants by a simple and relatively rapid chemical precipitation process from HCl solution by using 1,10-phenanthroline as a selective precipitating agent, (ii) determination of the decontamination ratios of the precipitated radiotellurium from the other radiocontaminants (by radiometric analysis), (iii) characterization of the formed precipitate, and (iv) recovery of the precipitated tellurium in a chemical form suitable for irradiation as a nuclear target.

## 2. Experimental

### 2.1. Chemicals and instruments

All chemicals used in the present work were of AR grade. Distilled water was used for preparation of solutions. A multichannel analyzer (MCA) of "Inspector 2000" model, Canberra Series, made in USA, coupled with a high-purity germanium coaxial detector (HPGe) of GX2518 model was used for γ-radioactivity identification and measurement. It was calibrated with a mixed source of $^{155}$Eu (86.5 and 105.3 keV), $^{57}$Co (122.1 and 136.5 keV), $^{137}$Cs (661.6 keV), $^{54}$Mn (834.8 keV), and $^{65}$Zn (1115.5 keV). The precipitate formed was characterized by (i) IR spectroscopy using Model 157-Bomem (Canada) and Model 6300-Jasco (Japan) FT-IR spectrometers, for mid-infrared range: 4000–400 cm$^{-1}$, and near-infrared range: 650–150 cm$^{-1}$, respectively, and (ii) thermal analysis using Shimadzu DTG-60H instrument for simultaneous DTA and TGA.