



Determination of trace selenium in high purity tellurium by hydride generation atomic fluorescence spectrometry after solid phase extraction of a diaminobenzidine-selenium chelate

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ABSTRACT

Macroporous adsorption resin was used as the sorbent for solid phase extraction and determination of the trace Se content in high purity tellurium prior to hydride generation atomic fluorescence spectrometry analysis. Selenium was converted into an organic Se chelate using 3,3'-diaminobenzidine and was separated from the tellurium matrix by solid phase extraction. The resin was packed as a column for solid phase extraction. Under optimum conditions, trace Se can be quantitatively extracted and the tellurium matrix can be removed. The Se in the eluate was determined by hydride generation atomic fluorescence spectrometry. The limit of detection (3σ) of this method was 0.22 ng g^{-1} and the relative standard deviation (RSD, $n = 5$) ranged from 2.0 to 2.5% for the three investigated tellurium samples. The proposed method was successfully applied for the determination of the trace Se content in high purity tellurium samples.

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1. Introduction

High purity tellurium is widely used in photovoltaic cells [1], semiconductors [2] and radiation detectors [3]. Even trace amounts of impurities can have detrimental effects on the properties of these devices. Selenium exhibits similar chemical and physical properties to those of tellurium, making it difficult to separate Se from tellurium. As a result, Se is an important impurity in high purity tellurium [4–7]. Therefore, it is necessary to determine the amount of trace Se in high purity tellurium. Ma et al. [8] attempted to directly determine the amount of trace Se by hydride generation atomic fluorescence spectrometry (HG-AFS) using citric acid to eliminate the interference of the tellurium matrix. In fact, high tellurium matrix will lead to serious interference to trace Se in the hydride generation process. Therefore, separation and preconcentration are required prior to the determination of the trace Se. Various separation or preconcentration methods, such as coprecipitation [9], precipitation [10] and vacuum distillation [11], have been developed for routine analysis of impurities in high purity tellurium. Nevertheless, only two solvent extraction methods [12,13] are available for trace Se separation. Because large amounts of hazardous reagents were used for solvent extraction methods and are not environmentally friendly.

Solid phase extraction (SPE) is widely applied for separation or preconcentration of trace elements because of its well-known advantages, such as low solvent consumption, potential for automation, high

reproducible recovery, and fast phase separation. SPE methods for separation of Se have been reviewed in two articles in the literature [14,15]. Two SPE strategies were used for Se separation on the basis of the different sorption materials. The first material adsorbs inorganic Se species [16,17]. The second material adsorbs Se chelate. In the latter case, inorganic Se species need to be converted into Se chelate using chelation agents, such as 2,3-diaminonaphthalene (DAN) [18,19] and ammonium pyrrolidine dithiocarbamate (APDC) [20–23], followed by SPE. APDC is a non-specific and multi-element complex reagent for trace elements [24] that is unsuitable for the selective separation of trace Se in tellurium [21]. DAB is a specific chelation agent for Se [25,26]. Although DAN is also a selective chelation agent for Se, it requires a longer time and higher temperature of reaction with Se than DAB [27,28].

Various sorbents, such as polymers and silica gel-based sorbents, activated carbon, metal oxides and hydroxides, have been used for solid phase extraction of Se [15]. D101 resin is a MAR of styrene copolymer and that is highly cross-linked, is non-ionic (non-functionalized), and has a relatively larger surface area. It is suitable for retaining hydrophobic compounds, especially aromatics [29].

The aim of this work is to develop a selective and sensitive method to determine trace Se in high purity tellurium. For this purpose, SPE was used to separate trace Se in a tellurium matrix. DAB was selected as the chelation agent for the conversion of Se (IV) into an aromatic chelate (diphenylpiaszelenol) [30]. D101 resin was used as the sorbent. Furthermore, HG-AFS was used to determine the Se content in the eluate because of its unmatched sensitivity, high analyte transport efficiency, and high selectivity [31,32].

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2. Experiments

2.1. Instruments

An AFS-930 atomic fluorescence spectrometer (Beijing Titan Instrumentals Co. Ltd., Beijing, China) equipped with a sequential injection hydride generation sampling system was used to determine the trace Se content. The AFS-930 specifications are given in Table 1. An Elan DRC-e inductively coupled plasma mass spectrometer (ICP-MS) (Perkin-Elmer, Concord, Canada) was used to determine the high concentration ($\mu\text{g mL}^{-1}$ level) of Se in the evaluation experiment of the resin capacity and the trace tellurium matrix remaining in eluate.

A digital temperature controllable hot plate (EH35B, Beijing LabTech Instruments Co. Ltd., Beijing, China) was used for sample treatment. A pH meter (DP PHS-25A, Shanghai Dapu Instrument Co. Ltd., China) with glass electrode was used for the pH adjustment. An electronic thermostat water bath (SHA-CA, Changzhou Putian Instrument Ltd., Changzhou, China) was used to control the reaction temperature of Se with DAB. One-hundred-millilitre PTFE beakers with a cover were used for the reduction of Se (VI). A quartz column with a 50-mL reservoir and a stopcock (250 mm length and 8 mm inner diameter) was used as the SPE column.

2.2. Reagents and solution

All reagents were prepared from analytical grade materials and all solutions were prepared with ultrapure water (18.2 M Ω cm). D101 macroporous adsorption resin (polystyrene type) was purchased from Tianjin Guanfu Fine Chemical Institute (Tianjin, China). The Se (IV) stock solution (1000 $\mu\text{g mL}^{-1}$) was prepared using sodium selenite ($\text{Na}_2\text{SeO}_3 \cdot 5\text{H}_2\text{O}$, 99%, Aladdin, China) and standard solutions of Se were prepared by diluting the stock solution. The following solutions were prepared: ethylene diamine tetraacetic acid (EDTA) (0.05 mol L $^{-1}$); citric acid (40%, m/v) in water; diaminobenzidine dihydrochloride (DAB) (0.5%, m/v, prepared daily) in water; formic acid (5%, v/v) in water; an ethanol solution containing 5% (v/v) nitric acid as the eluent; hydrogen peroxide (30%, v/v); Fe (III) solution (10 mg mL $^{-1}$) in a nitric acid solution for the Fenton reaction, and HCl (6 mol L $^{-1}$) for the reduction of Se (VI) to Se (IV). A KBH_4 (1.0%, m/v) solution was used as a reductant and was prepared daily by dissolving in a 0.5% (m/v) KOH solution, and HCl (5%, v/v) was used as the carrier for HG-AFS.

2.3. Column preparation

A small plug of glass wool was placed on top of the stopcock of the quartz column as a support for the resin. One gramme of the D101 resin suspended in water was slurry-packed in the quartz column. Another glass wool layer was placed in the upper part of the column. The resin was first soaked with 10 times its volume of 95% ethanol (v/v) for 24 h, and was then rinsed with 95% ethanol (v/v) (flow rate, 1.0 mL min $^{-1}$) until the effluent was no longer turbid when mixed with 5 times the amount of water. Finally, the resin was successively

rinsed with 5% (v/v) HCl and water until there was no ethanol odour and the effluent was neutral and ready for solid phase extraction.

2.4. Sample preparation

According to the sample content, approximately 0.5–1.0 g high purity tellurium powder were accurately weighted (to the nearest 0.002 g) in a 100-mL quartz beaker. Fifteen millilitres of 30% (v/v) nitric acid was used to dissolve the sample on a controllable hot plate at 120 °C until the sample was completely decomposed. The sample solution was subsequently evaporated to near dryness. A small amount of water was used to rinse the beaker wall and the solution was evaporated to near dryness again. Ten millilitres of 6 mol L $^{-1}$ HCl was added to convert Se into its quadrivalent form [33]. The beaker was covered with a watch glass, and the solution was heated to 100 °C for about 10 min to dissolve the residue. After the solution was cooled, 5 mL of 0.05 mol L $^{-1}$ EDTA and 10 mL of 40% (m/v) citric acid were added. Then the solution was adjusted to pH 2.8 ± 0.1 [12] using a pH meter with concentrated and diluted ammonia solutions, respectively. Five millilitres of a 0.5% (m/v) 3:3'-diaminobenzidine dihydrochloride solution was added. The solution was placed in a 50 °C water bath for 50 min. The solution was then cooled to room temperature for solid phase extraction.

2.5. Solid phase extraction procedure

The abovementioned sample solution was allowed to pass through the D101 resin column at a flow rate of 3 mL min $^{-1}$. Then, the column was rinsed with 40 mL of 5% (v/v) formic acid (pH 2.8 ± 0.1). The adsorbed Se chelate in the resin was eluted with 30 mL of 5% (v/v) nitric acid in ethanol at a flow rate of 3 mL min $^{-1}$. The eluate was collected in a 100-mL Teflon beaker.

2.6. Determination of Se in eluate

The Se species in the eluate exist in an organic chelate form that is not favourable for hydride generation. To decompose the organic substances in the eluate, 2 mL of concentrated nitric acid, 2 mL of 30% (v/v) hydrogen peroxide and 0.4 mL of 10 g L $^{-1}$ Fe (III) were added to the eluate. The solution was then evaporated to near dryness. An additional 2 mL of hydrogen peroxide was then added. The solution was evaporated to near dryness again. The Fe (III) ion acted as the catalyst for the Fenton reaction, which is able to effectively break down organic substances [34]. Finally, 2 mL of 6 mol L $^{-1}$ HCl was added to dissolve the residue and reduce Se (VI) into Se (IV). The sample solution was diluted to 10 mL for HG-AFS analysis. An external calibration curve was established using a standard Se (IV) solution for calibration. Finally, the recoveries of Se under different conditions were obtained by comparing the concentration of Se in the eluate with the spiked concentration levels.

3. Results and discussion

3.1. Effect of adsorption pH

The effect of pH on the adsorption of the Se-DAB chelate on the D101 resin was investigated in the 2.3–8.0 pH range (Fig. 1). After the chelation reaction of Se and DAB, the solution was adjusted to the given pH with ammonia and formic acid. Meanwhile, the column was equilibrated with the corresponding pH buffer solution prior to loading the sample solution. The following steps were completed in accordance with the general procedure. The results showed that quantitative adsorption of the Se-DAB chelate was achieved in the 2.3–8.0 pH range. A pH range of 2.8 ± 0.1 was selected for further experiments; thus, the sample solution did not require an additional pH adjustment after the Se chelation reaction.

Table 1
HG-AFS instrument specifications.

PTM voltage (V)	300
Atomizer temperature (°C)	200
Atomizer height (mm)	10
Main lamp current (mA)	80
Auxiliary lamp current (mA)	30
Flow rate of carrier gas (Ar) (mL min $^{-1}$)	400
Flow rate of shield gas (Ar) (mL min $^{-1}$)	800
Signal processing mode	Peak area
Carrier solution (HCl)	5% (v/v)
Reducing solution (KBH_4)	1% (m/v, in 0.5% NaOH solution)

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