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Review Article

A REVIEW ON THE CHEMICAL ANALYSIS OF TELLURIUM AND ITS REMOVAL FROM REAL AND BIOLOGICAL SAMPLES

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ABSTRACT

Tellurium demonstrates properties similar to those of elements known to be toxic to humans, and has applications in industrial processes that are rapidly growing in importance and scale. It is also considered to be extremely toxic, and clinical manifestations of toxicity are observed at very low concentrations. Therefore, there are several methods interested in determination and removal of tellurium in its environmental and biological samples. In this review, we will focus on the protocols that deal with determination, speciation and removal of tellurium in real and biological samples. This includes volumetric, spectral, chromatographic and potentiometric methods, etc. This review also highlights ongoing research activities and presents the challenges that should be addressed in the future.

Keywords: Tellurium, Analysis, Removal, Speciation.

1. INTRODUCTION

Tellurium (Te) is a metalloid element belonging to the VIA group of the periodic table that shares some chemical properties with elements such as oxygen, sulfur, and selenium[1]. Tellurium was discovered in 1782 and was so called from the Latin word for earth, *tellus.* The abundance of tellurium in earth's crust is 0.01 ppm .It occurs mainly as tellurides with gold, silver, lead, and bismuth[2]. Tellurium occurs naturally in a number of inorganic forms, including telluride, tellurite and tellurate.

Although tellurium is not widely used, it is required in a number of important industrial applications[3]. Together with elements, such as As, Sb, and Se, it plays a particular role in the manufacturing of semiconductors and other electronic components. It is a component of special alloys, where it improves hardness and resistance to corrosion, and also of certain glasses. Tellurium compounds are used in the rubber industries, the manufacture of batteries, and are found in fairly large amounts in the human body[4,5]. Tellurium improves the machinability of copper and stainless steel and its addition to lead decreases the corrosive action of sulfuric acid on lead and improves its hardness[6]. New applications include a germanium-antimony-tellurium compound used for optical storage on digital video discs[7]. There is some interest in the use of radioactive tellurium to treat thyroid cancer, and Te-containing antitumor agents have been developed[8].

Although tellurium is not an essential element and is relatively rare in the environment, it is also considered to be extremely toxic, and clinical manifestations of toxicity are observed at very low concentrations[2].

Tellurium can be accumulated mainly in the kidneys, heart, liver, spleen, bone, and lung and its threshold should not excess 2.5 mg/kg[2], if exceed it could induce the degeneracy of the liver and kidneys[9]. In humans, tellurium is partly metabolized into dimethyl telluride, $(CH_3)_2Te$ gas which is a characteristic feature of exposure, and gives a pungent garlic-like odor to breath, excreta, and the viscera. The main target sites for Te toxicity are the kidney, nervous system, skin, and the fetus (hydrocephalus). The emission of inorganic tellurium compounds in the environment may create serious problems due to the acute and chronic toxicity of this element. The most obvious sign of Te exposure is the garlic odor and other mild symptoms include dry mouth, metallic taste,

and somnolence[10]. Furthermore, tellurium's toxicity, bioavailability and environmental transport mechanism highly depend on its chemical form and oxidation state. For example, tellurite is 10 times more toxic than tellurate[11]. The Oxyanions of tellurium, particularly tellurite (TeO_3^{2-} or Te(IV), are highly toxic for most microorganisms[12]. The main reason for metal toxicity is oxidative damage of proteins, DNA, and lipids. It has been proposed that this toxicity results from an ability of Te(IV) to act as a strong oxidizing agent[13], as Te(IV) directly interacts with and oxidizes cellular thiol groups[14,15]. Te(IV) has also been implicated in redox reactions involving the respiratory chain[16]. Recent reports favor an oxidative mode of action; Te(IV) increases the production of reactive oxygen species (ROS) *in vitro* as well as *in vivo*, and increases protein carbonylation[17].

It has long been known that different bacterial species vary in their tolerance to Te(IV), which has prompted the inclusion of TeO_3^{2-} in a variety of selective bacteriological tests[18]. However, tellurite-resistant bacteria do exist in nature and they often reduce tellurite to its elemental less toxic form Te^0 that is accumulated as black deposits inside the cell[19,20]. Resistant cells grow as black colonies on relatively high concentrations of TeO_3^{2-}.

Tellurite has been used for 80 years for the isolation of pathogens including *Corynebacterium diphtheriae*, *Staphylococcus aureus*, *Vibrio cholerae* and *Shigella* spp. [21,22]. Also tellurite is used for selection of verocytotoxigenic *Escherichia coli* 0157, where tellurite allow the growth of VT^+ *E. coli* 0157 and *Shigella sonnei* but partially or completely inhibited the growth of other strains of *E. coli* [23].

Thus, Te demonstrates properties similar to those of elements known to be toxic to humans, and has applications in industrial processes that are rapidly growing in importance and scale. It is relevant, therefore, to consider the Te physiology, toxicity, and methods for monitoring the element in biological and environmental specimens. For these reasons, analytical chemists have paid special attention to development methods for tellurium speciation analysis. Several analytical techniques have been applied for speciation of tellurium in real and biological samples. The most important are volumetric, spectral, chromatographic and potentiometric methods, these methods are described below.

2. VOLUMETRIC METHODS

Masson[24] studied a procedure for titration of selenite, tellurite and arsenate with silver nitrate, and its application in the determination of organic and organometallic compounds. Decomposition is carried out either by oxygen-flask combustion or by wet digestion with sulphuric and nitric acids. Pure silver wire was used as an indicator electrode. The analysis is completed by argentometric titration, at pH 8-8.5, of the selenite, tellurite or arsenate formed. Halogens can often be determined simultaneously, and procedures are given for dealing with metal-containing compounds. Results obtained are within the usual limits for microanalysis (+0.3%).

A direct cerimetric method for determination Te(IV) was developed by Dindi and Reddy[25]. The sample (containing 10 to 100 mg of Te) was mixed with H_2SO_4 (to give a 1 to 5 mol L^{-1} when diluted to 50 mL) and with $RuCl_3$ as catalyst to give a mole ratio of Te(IV) to Ru of ≥ 800:1. The mixture was heated to ~100 ºC and titrated slowly with stirring with a standard solution of $Ce(NH_4)_4(SO_4)_4$. The end-point was the appearance of the first persistent pale yellow colour. The method can also be carried out in 1 to 2 mol L^{-1} HNO_3 with $Ce(NH_4)_2(NO_3)_6$. The coefficient of variation (n = 10) obtained with the use of H_2SO_4 or HNO_3 were 0.21 and 0.15%, respectively. No interference occurred in either media from Cu(II), AgI and Cl^- up to 2.5, 60 and 10 mmol L^{-1}, respectively, and Pb(II) does not interfere at up to 50 mmol L^{-1} in HNO_3 medium. All other ions that can be oxidized with Ce(IV) interfere. This method has been extended to the analysis of tellurate-tellurite mixtures by converting Te(VI) to Te(IV) by heating with conc. HCl.

Table 1: Literature data survey for volumetric determination of tellurium

Technique used	Form of Te	Linear range	LOD	Titrant	pH	Type of samples	Ref.
Argentometric titration	Te(VI) and Te(IV)	---	---	$AgNO_3$	8.0-8.5	Organic and organometallic compounds	[24]
Cerimetric titration	Te(IV)	----	----	$Ce(NH_4)_4(SO_4)_4$	Acidic	Tellurate-tellurite mixture	[25]

3. SPECTRAL METHODS

Spectrophotometric method for determination of tellurium as iodotellurite complex was studied by Johnson and Kewan[26]. Iodotellurite complex is soluble, highly colored and shows absorption maxima at 335 and 285 nm. In mixtures of 0.15 to 0.40 mol L^{-1} hydrochloric acid and 0.25 to 0.40 mol L^{-1} potassium iodide, the absorption of the iodotellurite complex at 335 nm follows Beer's law in the range 0.2 to 2 µg $mL^{-1}$of tellurium. Errors from the side reaction involving iodine formation by air oxidation of iodide are avoided by using low-actinic glassware and by making readings soon after mixing the reagents. Bismuth also forms a colored iodo complex and must be removed. Selenite is reduced to colloidal selenium by the iodide. Fe(III) and Cu(II) ions oxidize iodide and thus interfere. Convenient separations from each of these are described. Nitrate, sulfate, mercury(II), cadmium, zinc, nickel(II), and cobalt(II) ions do not interfere. This method can be used in metallurgy, biological science, and industrial hygiene.

Walper et al.[27] developed a method for the quantitative determination of tellurite in bacteriological culture medium depending on thioacetamide. Tellurite can be reduced to elemental tellurium with the action of hydrogen sulfide and readily forms colloidal solutions. Thioactamide was used as a convenient source of hydrogen sulfide. The tellurite solutions after reduction were measured spectrophotometry at wavelength 350 nm. the results obey Beer's law is the range 1-10 µg mL^{-1} of tellurite. The coefficient of variation for this method was 1.7%; the percentage error was 3.0.

Thompson et al.[28] studied a procedure for the simultaneous determination of arsenic, antimony, bismuth, selenium and tellurium by generation of their gaseous hydrides and introduction of these hydrides into an inductively coupled plasma source where the atomic line emission from the elements is detected. The effect of different plasma-torch assemblies on the detection limits attainable has been studied and suitable compromise conditions have been established for the simultaneous generation of the hydrides of the analyte elements and their determination by optical emission spectrometry in the plasma source. Detection limits of 1 ng mL^{-1} or below for aqueous solutions of the elements studied have been obtained under the conditions employed to allow their determination in samples of interest in geochemistry.

A method for determination of trace As(III) and As(V), Sb(III) and Sb(V), Se(IV) and Se(VI), Te(IV) and Te(V1) in water by atomic-absorption spectrophotometry after separation and enrichment with "thiol cotton" and hydride generation has been established[29]. The sorption behavior of various oxidation states of arsenic, antimony, selenium and tellurium, and the conditions of quantitative sorption and desorption of these species were studied. The procedures for reducing species from higher oxidation states were optimized. Interferences from other species and their elimination were investigated. The selectivity of the procedure for the determination of species in higher and lower oxidation states was examined. This method was applied in determination of arsenic, antimony, selenium and tellurium in water, in the range from pg mL^{-1} to ng mL^{-1}. The recoveries for added spikes were in the range 90-110%, with coefficients of variation in the range 3-8%.

For the quantitative determination of tellurite in biological media, Turner et al.[30] described a method depending on the use of diethyldithiocarbamate. In earlier protocols, diethyldithiocarbamate reacted with tellurite and the resulting complex was extracted into organic solvents before spectrophotometric determination. In this study, Tellurite containing fermentation broth (100 µL) was mixed with 0.5 mol L^{-1} Tris-HCl (pH 7.0, 300 µL) and 10 mmol L^{-1} diethyldithiocarbamate (100 µL) and diluted to 500 µL with H_2O to form a yellow colloidal solution. The absorbance of the aqueous yellow solution was measured at 340 nm. Beer`s law was obeyed from 1 to 50 µg mL^{-1} (4 to 200 µmol L^{-1}) of tellurite (ε= 5080). Decrease of absorbance caused by solution degradation was 27% after 2 hours, absorbance should, therefore, be measured in 4 hours. The method was applied to *Escherichia coli*.

The occupational exposure to Te was studied by Tylor [2]. Hydride generation atomic absorption spectrometry provides accurate, precise, and timely measurements results. The measurement occurred at 214.3 nm. Animal studies suggest that up to 25% of orally administered tellurium is absorbed in the gut. There is a biphasic elimination from the circulation with loss of about 50% within a short period, $t_{1/2}$= 0.81 d, and slower elimination of the residual Te, $t_{1/2}$ = 12.9 d. The results showed that the minimum fatal dose of Na_2TeO_3 is 2.25-2.50 mg/kg while Na_2TeO_4 is 20-30 mg/kg, so tellurite is more toxic approx 10-fold than tellurate.

Najafi et al.[31] developed a method for speciation and determination of Te(IV) and Te(VI). The method depends on dispersive liquid-liquid microextraction combined with electrothermal atomic absorption spectrometry

using palladium as permanent modifier. Under acidic conditions (pH 1), only Te(IV) can form a complex with ammonium pyrrolidine dithiocarbamate (APDC) and, therefore, be extracted into fine droplets of carbon tetrachloride (extraction solvent) which are dispersed with ethanol into the water sample solution. After centrifugation, Te(IV) was determined in the sedimented organic phase while Te(VI) remained in the aqueous phase. Total inorganic tellurium was determined after the reduction of the Te(VI) to Te(IV). Te(VI) was calculated as the difference between the measured total inorganic tellurium and Te(IV) content. The effective parameters for improving the efficiency of microextraction process were investigated by using experimental and central composite designs. Under optimal conditions the enrichment factor was 125 and the calibration graph was linear in the range of 0.015-1.0 ng mL^{-1} with detection limit and characteristic mass of 0.004 ng mL^{-1} and 0.033 pg, respectively. The relative standard deviation for 0.5 ng mL^{-1} of tellurium measurement was 3.6% (n=6) at ash and atomization temperature, 900 and 2600 $^{\circ}C$, respectively. The recoveries of spiked Te(IV) and Te(VI) to the environmental water samples were 89.6-101.3% and 96.6-99.1%, respectively. The accuracy is also evaluated by applying the proposed method to certified reference material (NIST SRM 1643e), for which the result was in a good agreement with the certified values reported for this CRM (95% confidence level).

Molina et al.[32] described a chemical method for tellurite quantification. The procedure is based on the $NaBH_4$ mediated reduction of TeO_3^{2-} followed by the spectrophotometric determination of elemental tellurium in solution. The method was applied in culture media and showed reproducible results, stable at different pH values, and exhibits linearity over a broad range of tellurite concentrations.

Hydride generation was inducted into the chemiluminescence for the determination of tellurium (IV) by Luo et al.[33]. The experiment exhibited that the strong chemiluminescence emission can be obtained during the reaction between hydrogen telluride and luminol in basic medium, and a novel sensitive hydride generation chemiluminescence (HG-CL) methodology for the determination of tellurium was proposed. Under the optimized conditions, the linear range of CL intensity versus concentration of tellurium (IV) was 10–200 µg L^{-1}, with a correlation coefficient (r^2) of 0.997 and a limit of detection (S/N=3) of 2 µg L^{-1}. The results showed that the method provided superior performance with respect to tolerance to various coexisting ions such as Mg^{2+}, Ca^{2+}, Fe^{3+}, Zn^{2+}, Pb^{2+}, As^{3+}, Ge^{2+}, and Hg^{2+}. The proposed method can be applied in detecting tellurium in environmental and biological samples.

Zhang et al.[34] proposed a simultaneous multi-channel hydride generation atomic fluorescence spectrometry (HG-AFS) method for determination of total arsenic (As), total bismuth (Bi), total tellurium (Te) and total selenium (Se) in tea leaves. The operating parameters of self-made multi-channel HG-AFS were optimized, including negative high voltage of photomultiplier tube (PMT), the flow rates of carrier and shield gas, observation height and lamp currents. The conditions of hydride generation for As, Bi, Te and Se were studied in details. Under optimal conditions, the method detection limits (MDL) for As, Bi, Te and Se in tea leaves were 0.0152, 0.0080, 0.0022 and 0.0068 µg/g, respectively. The proposed method was applied to the simultaneous determination of As, Bi, Te and Se in various tea leaves and the spike recoveries were in the range of 90-103%. The accuracy of method was validated by analyzing a tea certified reference material. The obtained values were consistent with the certified ones.

An electrolysis process for the preconcentration of selenium (IV) and tellurium (IV) is reported[35]. Where, Electrodeposition technique used for sample preparation in X-ray fluorescence spectrometry with advantages of low cost, high sensitivity, and minimal interferences. The analytes were electrodeposited on a polished substrate of high purity copper, which was rinsed with deionized water, dried, and directly analyzed by x-ray fluorescence. The influence of concomitant metal ions, electrodeposition voltage, pH, and electrolyte concentration on the deposition efficiency was investigated. The reproducibility of determination was 2.4% for selenium (IV) and 1.9% for tellurium (IV) (n = 7). The detection limits for the measurement of selenium (IV) and tellurium (IV) were 0.44 and 0.57 µg L^{-1}, respectively. The method was validated by analyzing certified reference materials and applied to the determination of trace selenium and tellurium in environmental water samples with satisfactory results.

Table 2: Literature data survey on spectral methods for tellurium determination

Technique used	Form of Te	Linear range	LOD	Reagent	pH	λ_{max} (nm)	ε	Type of samples	Ref
Spectrophotometry	Te(IV)	0.2-2 µg mL^{-1}	---	KI	Acidic (HCl)	335	--	Pure solution	[26]
Spectrophotometry	Te(IV)	1-10 µg mL^{-1}	--	Thioacetamide	Neutral	350	--	Bacteriological culture medium	[27]
ICP-emission spectrometry									[28]
Hydride generation-AAS	Te(IV) and Te(VI)	pg mL^{-1} to ng mL^{-1}	0.008 ng mL^{-1}	Thiol cotton (separation) potassium borohydride	3.0	214.2	---	Water	[29]
Spectrophotometry	Te(IV)	1 to 50 µg mL^{-1}	---	Diethyldithiocarbamate	7.0	340	5080	Biological media (*E coli*)	[30]
Hydride generation-AAS	Te(IV) and Te(VI)	---	---	Sodium borohydride	Alkaline NaOH	214.3	---	Biological samples(blood, urine membranes)	[2]
Dispersive liquid–liquid extraction - AAS	Te(IV) and Te(VI)	0.015-1 ng mL^{-1}	0.004 ng mL^{-1}	Palladium as permanent modifier Sodium borohydride	1.0	214.3	---	Water samples	[31]
Spectrophotometry	Te(IV)	1-200 µg mL^{-1}	---	$NaBH_4$	pH depentant	500	---	Culture media	[32]
Hydride generation-chemiluminescence (HG-CL)	Te(IV)	10-200 µg L^{-1}	2 µg L^{-1}	KBH_4 Luminol	11.5	---	---	Environmental and biological samples	[33]
Hydride generation atomic fluorescence spectrometry (HG-AFS)	Te(IV)	0.1-10 ng mL^{-1}	0.0022 µg/g	Potassium tetrahydroborate KBH_4	---	---	---	Tea leaves	[34]
X-Ray Fluorescence	Te(IV)	---	0.57 µg L^{-1}	Electrodeposition of tellurium on a polished substrate of copper	---	----	---	Environmental water samples	[35]

4. SEPARATION AND CHROMATOGRAPHIC METHODS

Lederer et al.[36] examined the eluation of tellurite and selenite on paper impregnated with dowex-50 in presence of numerous acids. It was possible to relate R_M value [$R_M = \log(1/R_F-1)$] with the pH of the eluting acid. This relationship can also be used for the determination of the charge on ion.

The separation of tellurite, selenite and sulfite ions by anion exchange resins was carried out by Iguch[37]. The distribution coefficients of tellurite, selenite and sulfite in neutral nitrate solution and alkaline hydroxide solution were measured and the results suggest the presence of an acid salt ion HXO_3^- in neutral solution. By the addition of ammonia to the eluant solution, the distribution coefficient of these salts in nitrate solution was increased, but in hydroxide solution it was decreased. However, by the addition of ethanol to the eluant solution that containing nitrate and ammonia, the distribution coefficient of sulfite increased, but that of tellurite and selenite remained constant even up to 75%. On the contrary, on the addition of ethanol to the hydroxide solution, the distribution coefficient of sulfite decreased, while that of tellurite and selenite increased at first and then decreased. This assay provided a complete separation of tellurite, selenite by the anion exchange using an alkaline eluant.

Yoshida and Hida[38] described an isotachophoresis method for separation of selenate, selenite, tellurate and tellurite ions. The effect of the pH of the leading electrolyte and the type of counter-ion on the separation of the cited anions was examined. Ion-pairing equilibria between the anions and the Ni(II)-1,10-phenanthroline and Co(III)-ethylenediamine complexes were the basis of the separation.

Selenite and tellurite were separated using extraction chromatography by Hu et al.[39]. The separation depends on the use of N263 (a commercial C8 to C10 trialkylmethylammonium chloride) as stationary liquid phase and aqueous sodium tartrate as mobile phase. With use of a column (10.4 cm x 8.4 mm) separation was achieved in 10 min. The SeO_3^{2-} content of some

samples was determined, and the general behavior of SeO_3^{2-} and TeO_3^{2-} was studied.

Fung and Lau[40] studied a new capillary electrophoresis (CE) procedure for simultaneous determination of ten oxoanions (CrO_4^{2-}, SeO_4^{2-}, MoO_4^{2-}, WO_4^{2-}, VO_4^{3-}, SeO_3^{2-}, AsO_4^{3-}, TeO_3^{2-}, TeO_4^{2-}, and AsO_3^{3-}) which were baseline-separated from each other and from the interfering UV absorbing anions (NO_3^- and NO_2^-) commonly found in environmental water samples. The new background electrolyte system developed contained 5 mmol L^{-1} potassium phosphate and 0.007 mmol L^{-1} octadecyltrimethyl-ammonium hydroxide, pH 11.2. The optimized working conditions were electrokinetic sampling at -5 kV for 10 s, running voltage at -15 kV with 5 µA current, and detection wavelength at 205 nm. No interference was observed for non-UV-absorbing anions and UV-absorbing anions up to 20 and 10 times higher concentrations, respectively. The analysis was fast, with a complete CE run within 6 min. Wide linear ranges (1-2000 µg L^{-1}), good repeatability in migration time (relative standard deviation RSD 0.55-2.8%), satisfactory precision in peak area (RSD 3.8-5.6%) and peak height (RSD 3.9-5.3%) measurement, and detection limits (1-25 µg L^{-1}) sufficiently sensitive to detect oxoanions found in environmental water samples. The reliability of the CE procedure developed had been established by recovery test and parallel method determination using atomic absoprtion spectrophotometry for real river water sample.

Optimization of the hyphenation between capillary zone electrophoresis (CZE) and inductively coupled plasma mass spectrometry (ICP-MS) was studied for the simultaneous determination of metalloid species in the environment[41]. Arsenic (arsenite, arsenate, monomethylarsonic acid, dimethylarsinic acid), selenium (selenite, selenate, selenomethionine, selenocystine), antimony (antimonite) and tellurium (tellurite, tellurate) species were simultaneously separated using a 75 µm i.d. fused silica capillary using either a chromate or a phosphate electrolyte. Different nebulizers were tested for introduction in the detector. A V-groove nebulizer (the Babington) and two concentric micronebulizers (the MCN-100 and the MicroMist) were studied in order to improve resolution, sensitivity and reproducibility. The optimization of CZE-ICP-MS interface operating parameters is discussed for each nebulizer-interface combination, and special attention is given to the position of the capillary inside the nebulizer. Different nebulizer gas and liquid sheath flow rates were studied in detail and they hardly affect electrophoretic resolution and peak width. The best analytical performance characteristics were obtained with the MicroMist nebulizer. Detection limits with this nebulizer were found to range between 6 and 58 µg L^{-1} depending on the species investigated using pressure injection and below 1 µg L^{-1} for most of the species with electromigrative injection. Analysis of soil extracts showed that it was possible to carry out this technique on real samples.

Ogra et al.[42] studied the urinary tellurium metabolite in rats administered sodium tellurite. In this study, the urinary Te metabolites (UTMs) in rats were detected by using HPLC-ICP-MS and electrospray ionization (ESI)-MS. To clear UTMs identification, two different chromatographic mechanisms, i.e., multi-mode gel filtration and cation exchange column, were employed. The major UTM detected after ingestion of tellurite was trimethyltelluronium, and no urinary sugar metabolites containing Te were detected despite the fact that the major urinary selenometabolite was a selenosugar (methyl-2-acetamido-2-deoxy-1-seleno-b-D-galactopyranoside). Interestingly, the ingestion of tellurite enhanced the excretion of selenometabolites in urine. These results suggest that Te is discretely metabolized from selenium (Se), an essential element belonging to the same group, although it affects the metabolism of Selenium in rats. Thus, the disturbance of Se metabolism, i.e., the induction of Se deficiency, may be one of the potential toxic effects of Te.

A simple capillary zone electrophoretic method for the determination of important oxyanions of selenium and tellurium and another Se-containing anion, selenocyanate, has been developed[43]. The method used direct UV absorption detection and allows determination of all the analytes in less than 6 min this assay has a lower detection limit compared with other methods involving complex biological matrices and with a wide linear range. The method was applied to live cultures of two different bacteria in two different growth media in time course experiments following the changes in metalloid-containing anion concentrations. The results showed that this method is a useful means in determination of these analytes in bacterial cultures.

Kuo and Jiang[44] described an ion chromatography-inductively coupled plasma mass spectrometric (IC–ICP–MS) method for the speciation of selenium and tellurium compounds namely selenite [Se(IV)], selenate [Se(VI)], Se-methylselenocysteine (MeSeCys), selenomethione (SeMet), tellurite [Te(IV)] and

tellurate [Te(VI)]. Chromatographic separation is performed in gradient elution mode using 0.5 mmol L^{-1} ammonium citrate in 2% methanol (pH 3.7) and 20 mmol L^{-1} ammonium citrate in 2% methanol (pH 8.0). The analyses are carried out using dynamic reaction cell (DRC) ICP-MS. The DRC conditions have also been optimized to obtain interference free measurements of $^{78}Se^{+}$ and $^{80}Se^{+}$ which are otherwise interfered by $^{38}Ar^{40}Ar^{+}$ and $^{40}Ar^{40}Ar^{+}$, respectively. The detection limits of the procedure are in the range 0.01–0.03 ng Se mL^{-1} and 0.01–0.08 ng Te mL^{-1}. The accuracy of the method has been verified by comparing the sum of the concentrations of individual species obtained by the present procedure with the total concentration of the elements in two NIST SRMs Whole Milk Powder RM 8435 and Rice Flour SRM 1568a. The selenium and tellurium species are extracted from milk powder and rice flour samples by using Protease XIV at 70 °C on a water bath for 30 min.

The speciation of inorganic tellurium species in seawater by inductively coupled plasma-MS (ICP-MS) following selective magnetic SPE (MSPE) separation was developed by Hu[45]. Within the pH range of 2-9, tellurite (Te(IV)) could be quantitatively adsorbed on c-mercapto-propyltrimethoxysilane (c-MPTMS) modified silicacoated magnetic nanoparticles (MNPs), while the tellurate (Te(VI)) remained in solution. Without filtration or centrifugation, these tellurite-loaded MNPs could be separated easily from the aqueous solution by simply applying external magnetic field. The Te(IV) adsorbed on the MNPs could be recovered quantitatively using a solution containing 2 mol L^{-1} HCl and 0.03 mol L^{-1} $K_2Cr_2O_7$. Te(VI) was reduced to Te(IV) by L-cysteine prior to the determination of total tellurium, and its assay was based on subtracting Te(IV) from total tellurium. The parameters affecting the separation were investigated systematically and the optimal separation conditions were established. Under the optimal conditions, the LOD obtained for Te(IV) was 0.079 ng L^{-1}, while the precision was 7.0% (C = 10 ng L^{-1}, n = 7). The proposed method was successfully applied to the speciation of inorganic tellurium in seawater.

Yang et al.[46] reported a method for tellurium determination in soil and plant samples using sector field inductively coupled plasma mass spectrometry (SF-ICP-MS). Soil and plant samples were digested using Aqua regia. After appropriate dilution, Te in soil and plant samples was directly analyzed without any separation and preconcentration. This sample preparation approach avoided to a maximum extent any contamination and loss of Te prior to the analysis. The developed analytical method was validated by the analysis of soil/sediment and plant reference materials. Satisfactory detection limits of 0.17 ng g^{-1} for soil and 0.02 ng g^{-1} for plant samples were achieved, which meant that this method was applicable to studying the soil-to-plant transfer factor of Te. This method can be used for the estimation of internal radiation dose of radioactive tellurium due to the Fukushima Daiichi Nuclear Power Plant accident. Determination of Co, Ni and Te in alcoholic beverages by photochemical vapor generation combined with inductively coupled plasma mass spectrometry was described[47]. Volatile species of Co, Ni and Te were liberated from mixed formic acid and acetic acid media following exposure to a UV source. Limits of quantification are 0.5, 0.1, and 0.4 µg L^{-1} for Co, Ni and Te, respectively, which correspond to improvements 9, 35, and 3 times more over those obtained with conventional pneumatic nebulization. Method accuracy was demonstrated by comparison of the determined Co, Ni and Te concentrations with those obtained following sample digestion and determination using conventional solution nebulization for sample introduction, resulting in good agreement at a 95% confidence level

Table 3: Literature data survey on separation and chromatographic methods for tellurium determination

Technique used	Form of Te	Linear range	LOD	Conditions of measurement	pH	Time of analysis	Type of samples	Ref
Ion exchange Paper chrom.	Te(IV)	---	---	Paper impregnated with) dowex-50 and HCl, HBr, HNO_3, H_2SO_4, $HClO_4$ as mobile phase	Acidic	---	Mixture of selenite and tellurite	[36]
Anion-Exchange Resins	Te(IV)	---	---	Dowex 1-X8, 100-to 200-mesh as anion exchange resin and $NaNO_3$ or NaOH as eluant	Alkaline	---	Mixture of sulfite, selenite and tellurite	[37]
Isotachophoresis	Te(IV) and Te(VI)	----	---	Ni(II)-1,10-phenanthroline and Co(III)-ethylenediamine (with leading electrolyte to form ion-pair) 0.01 M Cl^- + NH_3(leading electrolyte)	10.0	15 min.	Mixture of selenite, selenate, tellurite and tellurate	[38]
Reversed-phase extraction chromatography	Te(IV)	---	---	N263 (a commercial C8 to C10 trialkylmethyl-ammonium chloride) as stationary phase sodium tartrate as mobile phase	--	10 min	---	[39]
Capillary electrophoresis	Te(IV) and Te(VI)	1-2000 µg L^{-1}	1-25 µg L^{-1}	Electrokinetic sampling at -5 kV for 10 s, running voltage at -15 kV with 5 µA current, and detection wavelength at 205 nm	11.2	6 min	River water	[40]
Capillary zone electrophoresis (CZE)-(ICP MS)	Te(IV) and Te(VI)	---	6-58 µg L^{-1}	Fused silica capillary and chromate or a phosphate as electrolyte	---	---	Soil extract	[41]
(HPLC-ICP-MS)	Te(IV)	---	0.9 µg L^{-1}	The multi-mode gel filtration and cation exchange columns and ammonium acetate as eluant	6.5 and 8.0	---	Rats urine	[42]
Capillary zone electrophoresis	Te(IV) and Te(VI)	1.27-127 µg mL^{-1}	0.191 µg mL^{-1}	Fused silica capillary with separation voltage of -25 kV Detection by a photodiode detector at 220 nm	10.5	6	Bacterial cultures	[43]
(IC–ICP–MS)	Te(IV) and Te(VI)	---	0.01-0.08 ng L^{-1}	Gradient elution mode using ammonium acetate	3.7 and 8.0	≤12	Biological samples	[44]
Magnetic SPE followed by (ICP-MS)	Te(IV)	---	0.079 ng L^{-1}	c-mercapto-propyltrimethoxysilane modified silicacoated magnetic nanoparticles (MNPs)	2.0-9.0	---	Seawater	[45]
Sector field inductively coupled plasma mass spectrometry (SF-ICP-MS)	Te	---	0.17 ng g^{-1} (soil) 0.02 ng g^{-1} (plant)	Samples was directly analyzed without any separation and preconcentration	---	---	Soil and plant samples	[46]
Photochemical vapor generation with (ICP-MS)	Te	---	0.4 µg L^{-1}	Liberated volatile Te from formic acid and acetic acid media directed to UV source	---	---	Alcoholic beverages	[47]

5. ELECTROMETRIC METHODS

Issa and Awad[48] developed a potentiometric procedure for determination of quadrivalent tellurium Te(IV). Potassium permanganate oxidises quadrivalent tellurium in weakly alkaline solutions quantitatively at room temperature, without requiring precautions to be taken to exclude atmospheric oxygen. This method was applied to determine the iso-electric point of tellurium dioxide by estimating the tellurium content of buffer solutions saturated with the dioxide. The results obtained indicate that this point lies at pH 3.8. At this pH quantitative precipitation of tellurium dioxide takes place.

The analysis of tellurite-tellurate compounds by means of automatic amperometric and potentiometric titrations was carried out by Cornwell[49]. Tellurium (VI) is determined by the classical Bunsen method, where the chlorine is absorbed in an excess of potassium iodide and the liberated iodine is titrated automatically with sodium thiosulfate to an amperometric end-point. After fusion with potassium pyrosulfate, total tellurium and the associated metal ion are determined by automatic potentiometric redox and EDTA titrations, respectively.

Tellurium, as tellurite, can be determined with a fluoride-selective electrode[50] by means of an indirect procedure based on precipitation of tellurite with excess of lanthanum (III), followed by back-titration with standard fluoride. The end-point is located by using the Gran method, and the titrations are suitable for tellurite concentrations above 1 mmol L^{-1}.

Selig[51] described a potentiometric precipitation titrations of selenite, selenate, tellurite and tellurate. Selenite and tellurite were titrated with sodium diethyldithiocarbamate by using a silver selective electrode as sensor. TeO_3^2 only can be determined by potentiometric titration with hexadecylpyridinium chloride by using a PVC-dioctyl phthalate-coated graphite rod as sensor. Silver nitrate can be used to titrate TeO_3^{2-}, SeO_3^{2-} and TeO_4^{2-}. In aqueous solution SeO_3^{2-} could be titrated with $Pb(NO_3)_2$, whereas SeO_4^{2-} required a medium of 80% methanol. Neither SeO_3^{2-} and SeO_4^{2-} nor TeO_3^{2-} and TeO_4^{2-} could be titrated sequentially. Recoveries ranged from 94.74 to 100.84% with standard deviations from 0.1 to 0.71%.

Depending on differential pulse polarography, Kamal et al.[52] studied a method for determination of tellurate, tellurite, arsenite and metavanadate. Supporting electrolytes from a universal buffer series (Britton-Robinson) were used, with a SCE reference electrode, pulse amplitude of 100 mV and pulse rate of 2 mV s^{-1}. The effect of pH on peak potential is discussed. Detection limits and peak potential were 0.95 µmol L^{-1} at pH 8.3 and 4.45 µmol L^{-1} at pH 4.5 for TeO_4^{2-}, -1.13 V for TeO_3^{2-}, 4.5 µmol L^{-1} and -1.7 V for AsO_2^-, and -0.88 V for VO_3^-. Calibration graphs were rectilinear. Mixtures of TeO_4^{2-}-TeO_3^{2-}, TeO_4^{2-}-VO_3^-, and TeO_4^{2-}-AsO_2^- in the concentration range 0.95 to 50 µmol L^{-1} were analysed, with relative differences between actual and measured values in the range 1 to 10%.

Hassan[53] studied the polarographic behaviour of selenite and tellurite. Polarograms were obtained at 25 °C for 0.5 mmol L^{-1} of Se(IV) and Te(IV) (as their Na salts) in 1 mol L^{-1}-ammonium formate, acetate, tartrate, oxalate and benzoate solution and in the presence and absence of Triton X-100. The form of the polarograms and the mechanisms of the observed reductions are discussed. Plots of diffusion current versus concentration for the reduction wave of Se(IV) in all the salt solutions at -1.7 V versus its determination. Similarly, the concentration plots for Te(IV) were straight lines in the presence of Triton X-100. In binary mixtures of Se(IV) and Te(IV), the Te(IV) could be determined in 1 mol L^{-1} ammonium formate or oxalate containing 0.014 to 0.006% of surfactant or in 1 mol L^{-1} ammonium acetate, tartrate or benzoate containing 0.004% of surfactant. The limiting current was measured at -1.25 V versus SCE. The Se(IV) was determined in the presence of Te(IV) in acetate, tartrate and benzoate solution containing 0.004% of Triton X-100, with measurement at -1.7 V.

Tellurium ISE was prepared by Zareh and Amin[54] from a solid membrane comprising a mixture of $HgTeO_3/Hg_2Cl_2$ (1:1) with an internal contact of Hg metal in which a Pt wire was immersed. The electrode exhibited a Nernstian response of 29 mV/decade for the concentration range 10 µmol L^{-1} to 0.1 mol L^{-1} of tellurite. The working pH range was 3.5-10.5. The ISE was fairly selective to tellurite in the presence of most common ions and was used to determine tellurite in some binary mixtures of tellurite with chlorate, chromate, borate and arsenate. The mean recovery was 98-99% and the RSD were 0.19-0.51%.

The electrochemical behaviour of tellurium in 2.5 mol L^{-1} NaOH solution was studied for the recovery of tellurium from alkaline leach liquor of cemented Te using steady state polarization and cyclic voltammetry[55]. The deposition characteristics and the potential range for a stable deposit of tellurium were also investigated. The morphology of deposited Te in alkaline solution showed a very porous

nature and needle like radial growth. The potential range for stable electrodeposition was between -0.8 V and -0.95 V (vs Hg/HgO electrode), but electrowinning could be carried out at more negative potentials due to the disproportionation reaction of Te_2^{2-}. Laboratory-scale electrowinning experiments were performed under different operating voltages, temperatures and initial Te concentrations. The current efficiency was about 85±90% for 50% recovery and about 50±60% for 90% recovery. The purity of electrodeposited Te was higher than 99.95%.

Differential pulse voltammetric determination of trace Te(IV) was performed by Khoo and Ye [56]. Electropolymerization of 3,3-diaminobenzidine on a gold surface gave an adherent, stable film of poly(3,3-diaminobenzidine) (PDAB). This polymer film retained the complexational functionalities of its monomer, demonstrating preconcentration abilities for several ions, including Se(IV) and Te(IV). Continuous flow and flow injection methods were developed for the sensitive and selective determination of Te(IV). The optimized method for the continuous flow mode had a detection limit of 5.6×10^{-9} mol L^{-1} for 10 min preconcentration. Typical relative standard deviations for six consecutive determinations were 1.82 and 2.56% for Te(IV) concentrations of 1.0×10^{-6} and 5.0×10^{-8} mol L^{-1}, respectively (10 min preconcentration). The method was applied to the determination of Te(IV) in real samples.

A modified carbon paste electrode based on multi-walled carbon nanotube and Alizarin Red S acts as a chelating agent for tellurium(IV) ions by differential pulse stripping voltammetry, is described[57]. Under optimised operational conditions, the sensor exhibited linear behaviour in the range of 2.0–300 ng mL^{-1} (correlation coefficient: 0.9982) with a detection limit of 0.45 ng mL^{-1}. The results indicate that the sensor is sensitive and effective for the determination of tellurium in water samples and certified reference materials.

Biver et al[58] reported a method for determination of trace concentrations of Te(IV) and Te(VI) in surface waters by differential pulse cathodic stripping voltammetry. It is based on the proton reduction catalysed by the absorption of Te(IV) on the mercury electrode. Under optimum conditions (0.1 mol L^{-1} HCl), a detection limit of about 5 ng L^{-1} for a deposition time of 300 s is achieved. Organic matter does not represent a problem at low concentrations; higher concentrations are eliminated by adsorptive purification. Tellurium occurs primarily as Te(IV) and Te(VI) in natural water. Thus, determining total Te requires the reduction of Te(VI) that it is not electroactive. A number of reduction procedures have been carefully evaluated. However, there is an evidence that the addition of $TiCl_3$ to the acidified samples can reduce Te(VI) at trace levels to Te(IV) reliably and quantitatively. Therefore, the procedure described allows the direct determination of total Te and its redox speciation.

Table 4: Literature data survey on different electrometric methods for determination of tellurium

Technique used	Form of Te	Linear range	LOD	Measurements Conditions	pH	Type of samples	Ref
Potentiometric titration	Te(IV)	---	---	$KMnO_4$ oxidize Te(IV)	alkaline	Pure solution	[48]
Amperometric-potentiometric titrations	Te(IV) and Te(VI)	---	---	Sodium thiosulfate, potassium pyrosulfate and EDTA	---	Mixture of tellurite - tellurate	[49]
ISEs	Te(IV)	---	1.0 mmol L^{-1}	Flouride selective electrode	Neutral	Pure solution	[50]
Potentiometric argentometric titration	Te(IV) and Te(VI)	---	---	$AgNO_3$ or sodium diethyldithiocarbamate as a titrant	8.6 and 5.0	---	[51]
Differential pulse polarography	Te(IV) and Te(VI)	---	---	pulse amplitude (100 mV) pulse rate (2 mV s^{-1})	8.5 and 4.5	Tellurite-tellurate mixture	[52]
Polarography	Te(IV)	---	----	In presence of Triton X-100 at -1.25 V	---	Pure solutions,Te(IV) and Se(IV) mixture	[53]
ISE	Te(IV)	10^{-5}-10^{-1} mol L^{-1}	4.0×10^{-6} mol L^{-1}	$HgTeO_3/Hg_2Cl_2$ (1:1) as ion recognition	3.5-10.5	Binary mixtures of tellurite with chlorate, chromate, borate and arsenate	[54]
Cyclic voltammetry	Te(IV)	---	---	electrodeposition potential(-0.8 V to -0.95 V)	alkaline	alkaline leach liquor of cemented Te	[55]
Differential pulse voltammetric	Te(IV)	1×10^{-8}-2×10^{-6} mol L^{-1}	5.6×10^{-9} mol L^{-1}	film of poly(3,3-diaminobenzidine) (PDAB) on gold surface	4.0	Human urine, soil	[56]
Differential pulse stripping voltammetry	Te(IV)	2.0–300 ng mL^{-1}	0.45 ng mL^{-1}	multi-walled carbon nanotube and Alizarin Red S acts as a chelating agent	---	Water samples	[57]
Differential pulse stripping voltammetry.	Te(IV) and Te(VI)	Up to 10 µg L^{-1}	5 ng L^{-1}	proton reduction catalysed by the absorption of Te(IV) on the mercury electrode	Acidic	Surface water	[58]

6. BIOLOGICAL REMOVAL OF TELLURITE IONS

Cooper and Few[59] studied the uptake of potassium tellurite by a sensitive strain of *Escherichia coli*. By incorporating ^{127}Te into potassium tellurite (K_2TeO_3), the amount of the salt absorbed by a buffered suspension of a tellurite-sensitive strain of *Escherichia coli* could be calculated from a measure of the radioactivity remaining on the washed organisms. No radioactivity was lost by washing. The system responsible for tellurite uptake was unstable at low pH but did not deteriorate greatly over several hours at pH 5.5-7.3. The uptake of tellurite by the organisms increased with time, and was most rapid at pH 5.5 and 37 °C. This uptake has the properties of an enzyme action. Growth of *Escherichia coli* in a medium containing 0.05% glucose decreased the activity of the organisms, while the pH of the medium was scarcely affected. Heating to 100 °C for 20 min destroyed the enzymic system. The low uptake then obtained corresponded to a rapid combination with the cells which were saturated at fairly low tellurite concentrations.

Enzymic method for the determination of tellurite ions in aqueous media including blood plasma or serum is described[60]. The method based on the inhibition of Zn^{2+} - glycerophosphocholine phosphocholine phospho-diesterase by tellurite ions in the presence of tetramethylammonium ions. Assays were carried out at 20 °C in 0.1 mol L^{-1} glycine buffer of pH 10, containing 1 or 10 mmol L^{-1} tetramethylammonium chloride, 20 milliunits of the enzyme and 0.15 mmol L^{-1} p-nitrophenylphosphocholine substrate, with varying amounts of tellurite or serum. The nitrophenol formed was measured at 410 nm. The inhibition effect was expressed as a percentage of the control. Calibration graphs were linear when inhibition was plotted against log tellurite concentration for 0.25-4 µmol L^{-1} in the presence of 1 mmol L^{-1} tetramethylammonium chloride or for 50-800 nmol L^{-1} in the presence of 10 mmol L^{-1} tetramethylammonium chloride, with a detection limit of 50 nmol mL^{-1} tellurite. In the range 0.05-4 µmol mL^{-1} the RSD were 4-8.1%. There was no interference from phosphate, selenite, sulfite, arsenite or thiols. The method was used to study the removal of tellurites by thiols and the rate of uptake of tellurite by blood.

The uptake by light-grown cells of *Rhodobacter capsulatus* of the highly toxic metalloid oxyanion tellurite (TeO_3^{2-}) was examined[61]. tellurite is rapidly taken up by illuminated cells in a process which is inhibited by the protonophore carbonyl cyanide-p-tri£uoromethoxyphenyl-hydrazone (FCCP) and by the K^+/H^+ exchanger nigericin. Notably, the light-driven membrane potential (ΔΨ) is enhanced by K_2TeO_3 ≥200 µmol L^{-1}. Further, tellurite uptake is largely insensitive to valinomycin, strongly repressed by the sulfhydryl reagent N-ethylethylmaleimide (NEM) and competitively inhibited by phosphate. Tellurite is transported into cells by a Δ pH-dependent, non-electrogenic process which is likely to involve the phosphate transporter (PiT family).

Amoozegar et al.[62] isolated 49 strains of moderately halophilic bacteria from the salty environments of Iran, a Gram-positive coccus designated as strain QW6 showed high capacity in the removal of toxic oxyanions of tellurium in a wide range of culture medium factors including pH (5.5-10.5), temperature (25-45 °C), various salts including NaCl, KCl, and Na_2SO_4 (0.5-4 mol L^{-1}), selenooxyanions (2-10 mmol L^{-1}), and at different concentrations of potassium tellurite (0.5-1 mmol L^{-1}) under aerobic condition. Phenotypic characterization and phylogenetic analyses based on 16S rDNA sequence comparisons indicated that this strain was a member of the genus *Salinicoccus*. The maximum tellurite removal was exhibited in 1.5 mol L^{-1} NaCl at 35 °C, while the activity reduced by 53% and 47% at 25 and 45 °C, respectively. The optimum pH for removal activity was shown to be 7.5, with 90% and 83% reduced removal capacities at the two extreme values of 5.5 and 10, respectively. The impact of different concentrations of selenooxyanions (2-10 mmol L^{-1}) on tellurite removal by strain QW6 was evaluated. The ability of strain QW6 for the removal of tellurite in the presence of 6 mmol L^{-1} selenite increased by 25%. The concentration of toxic potassium tellurite in the supernatant of the bacterial culture medium decreased by 99% (from 0.5 to 0.005 mmol L^{-1}) after 6 days and the color of the medium changed to black due to the formation of less toxic elemental tellurium.

Among 148 bacterial isolates from two types of polluted water, strain STG-83 showed maximum oxyanion reduction and resistance ability[63]. Sequencing of the 16S rDNA gene of STG-83 showed that the strain is closely related to *Bacillus pumilus* and morphological and biochemical tests confirmed the result. The strain was nitrate negative, but it could reduce half of tellurite in solution containing 1.0 mmol L^{-1} concentration and completely reduced selenite and selenate in solutions containing 1.0 mmol L^{-1}. Both reduction to elemental form and volatilization occurred in

case of all oxyanions tested, according to hydride generation atomic absorption spectroscopy and proton induced X-ray emission analytical methods. The strain was able to tolerate remarkably high concentrations of selenite (640 mmol L^{-1}), selenate (320 mmol L^{-1}), and tellurite (1250 mol L^{-1}); and tolerance to tellurite increased in presence of selenite and selenate. Biochemical tests and zymogram of extracted culture solutions on gel electrophoresis showed that the strain was nitrate negative and therefore, nitrate did not interfere with reduction of other oxyanions. Thus, the strain opens up good opportunities for the bioremediation of polluted water in natural environment, since nitrate usually inhibits or decelerates reduction of the mentioned toxic oxyanions.

From an extreme environment, Antarctica, 123 tellurite-resistant bacteria were isolated[64], and six new tellurite-resistant and tellurite-reducing bacterial strains were characterized. These strains were identified according to their 16S rRNA gene sequence as *Staphylococcus hameolyticus*, *Staphylococcus sciuri*, *Acinetobacter haemolyticus*, *Pseudomonas lini*, and two strains of *Psychrobacter immobilis*. The isolates display tellurite-resistance about 35 to 500 fold higher than *Escherichia coli* (Te-sensitive organism), and a high level of tellurite reduction which might be interesting for an application in the field of bioremediation or nanoparticle biosynthesis.

CONCLUSION

This review describes several methods for the determination of tellurium in environmental and biological samples. Despite the significant progress, current methods still suffer from sample preconcentration, inappropriate detection limits and lack of speciation of tellurium and its acid forms in chemical environments. Although Te is not abundant in the earth crust, it is often found with other elements such as As, Sb, S and Se with similar chemical properties. Researchers are faced with several challenges for determining tellurium in the presence of chemical interferences and real samples at low cast by easily manipulation technique.

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