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PUBLICAÇÃO IEA N.º 371
Janeiro — 1975

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SÃO PAULO — BRASIL

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Publicação IEA Nº 371
Janeiro 1975

* IIIrd. Interamerican Radiochemistry Meeting 02 to 06 December, 1974 - S. Paulo, Brasil

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ABSTRACT

This paper reports the findings that tellurium ions form a soluble cationic complex with thiourea in acid medium The cationic tellurium thiourea species is strongly absorbed on a cationic ion exchanger The retention of tellurium on the resin enables many interesting separation schemes for tellurium from various ions With special interest the separation of iodine from tellurium was studied An efficient and convenient iodine 132 generator is described the radioiodine being eluted with water or 0 9 g l NaCl when desired

Introduction

There are in the literature many procedures described for the absorption of tellurium on anion exchange resins Kraus and Nelson[1] reported that Te(IV) is strongly absorbed on Dowex 1 anionic resin from hydrochloric acid Hicks et al[2] showed that Te(IV) and Te(VI) are strongly absorbed on Dowex 2 anion exchange resin while Attebury et al[3] and Aoki[4] have reported that tellurium can be absorbed from 3M HCl on an anion resin column and Schindewolf[5] also reported that tellurium can be absorbed on Dowex 1 resin from strong HCl Stronski and Rybakow[6] reported on the separation of radioactive traces of tellurium using Soviet procedure anion exchangers Wish[7] has quantitatively separated radioactive tellurium from a mixture of fission products and uranium by use of Dowex 2 resin and phosphoric acid solutions, tellurium being not absorbed

On the other hand only very few procedures are described for separation of tellurium using cationic ion exchangers Smith and Reynolds[8] using radioactive tracers have shown that Te(IV) in a 0 1M oxalic acid solution is not absorbed by Dowex 50 cationic resin and in the same study those authors showed that Te(IV) was only slightly absorbed on Dowex 1 anionic resin when a 0 1M oxalic acid solution was passed through the column

No procedure was found in the current literature describing the absorption of tellurium on cationic ion exchange resin

The separation chemistry involving tellurium and iodine is of great interest in radiochemistry No satisfactory method for separation of iodine from tellurium has been described using ion exchange resins Greene[9] stated that none of the known ion exchange resins tested were found to be at all effective to separate tellurium and iodine from each other in an efficient and rapid manner

This paper reports our findings that the cationic complex formed between Te(IV) and thiourea(tu) is quantitatively and strongly absorbed on a cationic ion exchanger This sorption makes it possible for many interesting separation of tellurium from various ions With special interest, we studied the separation of iodine from tellurium utilizing the absorption of its

cationic species formed with thiourea in acid medium on a cationic resin

Thiourea as a Complex Forming Agent

Thiourea forms complexes of the amine type with numerous heavy metals, most of which are colorless and some only slightly soluble, as for instance silver, mercury and thallium Some elements give colored products in acid solution e g bismuth (yellow) tellurium (yellow[10], osmium (red) and ruthenium (blue) Antimony gives a weak yellow color, and palladium a stronger one, while selenium (IV) is reduced to the element (red) and rhenium develops a yellow coloration in the presence of stannous chloride Thiourea reacts with a number of cations and anions to give color reactions of analytical importance Yoe and Overholser[11] have investigated many of these reactions The reaction of tellurium with thiourea is very sensitive, a yellow color appearing with as little as 1 part of tellurium in 500,000 parts of solution[12]

Some complexes formed with thiourea are cationic, like $Cu(tu)^{+}$. It is reported that osmium (VIII or IV) form a cationic species described as $Os(tu)^{+++}$ [13] and ruthenium (III and IV) is reported to form complexes of the $Ru(tu)^{++}$ type[14] Rhodium forms a cationic complex with thiourea as well A proposed separation of rhodium from iridium (IV) is based on the absorption of cationic rhodium thiourea complex on the cation exchange resin, Dowex 50W X8, the iridum thiourea complex being anionic passed through the column[15]

The extraction behavior of U, Fe, Cd, In, Zn, Cu, Co, Ni Mn, Cr, Ag, Bi, Pb, Tl, Ru, Pd, Pt Ir, Os Au, Hg, Sn and Sb with tri n octylamine diluent from HCl, HNO_3, H_2SO_4 and HI and the effect of thiourea as a complexing agent, giving rise to cationic species with several of the cited elements, was studied by Abrão[16] It was clear that the stable cationic metal thiourea were not extracted by the amine, the organic phase, similarly to anionic resins, extracting only anionic species

Thiourea Method For Tellurium

The intense yellow color produced in the reaction of tellurium (IV) with thiourea is useful for the colorimetric determination of tellurium in moderately strong acid solutions Nielsch and Giefer have studied the spectrophotometry of the tellurium thiourea complex in nitric acid[17], sulfuric and phosphoric acids[18] Tellurium (VI) reacts with thiourea in a few minutes at room temperature Tellurium (IV) react only in hot solutions Jilek and Vrestal[19] have developed a differential analysis for the two tellurium valence states using tellurium thiourea reaction at room temperature and at the boiling point

In the present paper, we report that the yellow complex formed in acidic medium between tellurium (IV) and thiourea is cationic in character and can be retained on a strong cationic ion exchanger Based on this findings, a procedure for the separation of tellurium from iodine is outlined

Ion-Exchange Separation of Iodine From Tellurium

The existing methods for the separation of iodine and tellurium are inadequate in terms

of time and labor for the radiochemist The separation presented in this paper is fast, simple, and is based upon the behavior of the cationic tellurium thiourea complex toward a cation exchange resin The cationic tellurium (IV) thiourea species is retained by the resin, whereas the iodide passes through the column In the case of radiotellurium, the generated radioiodine is simply eluted with water If desired, the tellurium (IV) thiourea complex can be eluted with 6M HCl at room temperature The tellurium iodine separation is quantitative

A Novel Radioiodine-132 Generator

The new method proposed here offer a contribution to the tellurium iodine separation chemistry, allowing to prepare a convenient iodine-132 generator The milking of radioiodine from tellurium is extremely simple A cationic ion exchanger serves as an iodine source, from which carrier free radioiodine can be milked quickly, and repeatedly at any desired moment

Two to three ml of strong cationic Dowex 50W X8, 50 100 mesh, ion exchanger is transferred as a slurry and packed into a glass column (8 mm I.D.), preparing a bed column of about 30 mm high The column is washed with five bed volumes of 1M HCl, and the excess of acid washed out with demineralized water

The sample used for the separation was 5-10 ml of carrier-free telluric acid (^{132}Te ^{132}I). To this acid solution, 1 ml of 25 g/l hidroxylamine hydrochloride was added and the solution warmed up to 80 90°C during 5 minutes If desired, 1 to 5 mg of tellurite(ate) can be added as carrier, before addition of the hydroxylamine After reduction of tellurium (VI) to tellurium (IV), 2 ml of aqueous 50 g/l thiourea are added After 5 minutes the solution is percolated through the exchanger at a flow rate of 2-3 ml per minute The colorless fraction of the effluent contains the iodide ion (carrier free ^{132}I) The tellurium (IV) thiourea complex is retained at the top of the column and when tellurite is used as carrier, a sharp yellow zone is shown After the sample solution had completely passed through, the column is washed with water until no hydrogen ion is detected in the effluent The column is ready to be milked

Radioiodine 132 can be milked with water at any desired time The column is exhaustively eluted with water and no leakage of tellurium was observed In some experiments, carrier-free iodine 132 was eluted with 9 g/l NaCl (for medical use), the elution being very successful, and no leakage of tellurium was observed as well Whether using tellurium as carrier or not, the radioiodine 132 obtained by the described procedure is radioactively pure, without contamination of tellurium 132, as confirmed by decay curves and half life determination

If for any purpose the elution of tellurium is desired, it can be accomplished with 6M HCl at room temperature

Discussion

Although anion exchange resin may be used to effect the chromatographic separation of tellurium and iodine from each other as anions, the separation is not satisfactory and elution time as well as the volume of eluant are large The novel procedure described here using the characteristic complex formed between tellurium (IV) and thiourea, a cationic species, strongly held by a cationic ion exchanger, represents a simplification to the tellurium-iodine sepatation chemistry If small amount of tellurite ion is used as carrier, the process can be visually

followed the sorption of tellurium (IV) thiourea complex resulting in a beautiful yellow ring on the top of the column. Whether tellurium is used as carrier or not, iodine is eluted as carrier free radioisotope.

The time lag between addition of thiourea to the sample as introduction of the solution into the column is necessary in order to allow sufficient time for the complete formation of the complex. The tellurium (IV) thiourea complex is formed rapidly, but tellurium (VI) thiourea complex, if formed, is attained more slowly at room temperature and probably need warming to speed up its formation (probably it is reduced to tellurium (IV) and complexed). We considered it preferable to add a few milligrams of hydroxylamine hydrochloride prior to thiourea addition, tellurium (VI) being reduced to tellurium (IV).

A variety of separation techniques, solvent extraction for instance, has been applied to problems of isolation and radiochemical purification of nuclides. The procedure recommended here is very fast, and with this technique it is easy to repeat the operation of elution several times without great expenditure of time.

The selective elution of short lived iodine 132 from the ion exchange column is very rapid from the relatively longlived tellurium 132 parent, which remained strongly held on the ion exchanger. If necessary the eluant (water or NaCl solution) can be forced by compressed air or by the action of a hypodermic syringe through the column to enchance the elution speed.

Finally, the cation exchange column separation of iodine from tellurium can be used as a good example of the rapid ion exchange separation method.

Conclusion

The radioiodine generator described here allows a clean radiochemical separation of iodine 132 from tellurium 132, the iodine being easily eluted with demineralized water or 9 g/l NaCl solution at sufficiently high activity.

The generator can be recommended for classroom demonstration on radiochemistry, nucleonics training and strongly recommended for medical use (the elution being easily done by the physician at the place of application).

The herein mentioned technique is being studied for the separation of tellurium 132 from fission products[20] and for the recovery of irradiated (old cooled, used, stocked solution) telluric acid or elemental tellurium for iodine 131 production[21].

Acknowledgement

The author is grateful to Dra. Constância Pagano Gonçalves da Silva, Head of Coordenadoria de Processamento de Material Radioativo, who supplied carrier free ^{132}Te solution.

RESUMÉ

Cette etude montre la nouveaute presente pour les ions de tellure de former complexes cationiques

solubles avec la thiourée en milieu acide. Les espèces cationiques tellure thiourée sont fortement retenues dans un échangeur cationique. La rétention du tellure dans la résine permet schemas intéressants de séparation du tellure et d'autres ions. On peut présenter un efficient simple et convenable générateur de iode 132 dans lequel le radioiode est élué avec d'eau ou avec une solution de NaCl 9 g/l suivant les besoins.

RESUMO

~~Este trabalho informa sobre o novo faro de~~ Íons de telúrio formam complexos catiônicos solúveis com tiouréia em meio ácido. As espécies catiônicas telúrio tiouréia são fortemente absorvidas num trocador catiônico. A retenção do telúrio na resina permite ~~interessantes~~ esquemas de separação do telúrio de outros íons. ~~Com esperança nisso~~ foi estudada a separação iodo telúrio. Descreve-se um ~~eficiente~~ simples e conveniente gerador de iodo 132 do qual o radioiodo é eluído com água ou com solução NaCl 9 g/l quando desejado.

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