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METHODOLOGY FOR THE DETERMINATION OF ENVIRONMENTAL ^{129}I AND ^{99}Tc

E. I. du Pont de Nemours and Company
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METHODOLOGY FOR THE DETERMINATION OF ENVIRONMENTAL ^{129}I AND ^{99}Tc*

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ABSTRACT: The Savannah River Laboratory is using existing techniques and developing new methodology to determine the environmental impact of the Savannah River Plant with regard to ^{129}I and ^{99}Tc. ^{129}I is determined by neutron activation after the method of Brauer. Activation products are quantified by γ-ray spectroscopy [Ge(Li)] following chemical isolation. ^{125}I is used as a yield tracer. ^{129}I amounts as low as 3.8 fCi can be determined with 30-minute counting times. An isotope dilution method for ^{99}Tc based on a three-stage surface ionization mass spectrometer is being developed. Its chemical isolation scheme ends with the Tc loaded on a single ion-exchange bead for enhanced mass-spectrometric sensitivity. ^{97}Tc will be used as a yield tracer. A lower limit of 0.2 fCi is sought. A modified method using liquid scintillation counting has determined ^{99}Tc in some aqueous samples. These methods have confirmed that ^{129}I and ^{99}Tc can be highly mobile in the aqueous environment, establishing the need for monitoring.

KEY WORDS: low-level analysis, ^{129}I, ^{99}Tc, neutron activation, gamma spectroscopy, environment, seepage basins, groundwater

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INTRODUCTION

Measurable quantities of anthropogenic ^{129}I and ^{99}Tc now exist in the world as a result of nuclear weapons testing, nuclear fuel reprocessing, and reactor operation. This results from high fission yield (6.3% for ^{99}Tc and 0.9% for ^{129}I from ^{235}U) and very long half-lives (2.12×10^5 and 1.7×10^7 years, respectively).

The biological behavior of radioiodine and its pathways to man have been thoroughly studied with regard to ^{131}I. Major pathways for thyroid dose by airborne radioiodine are consumption of milk by infants and milk and leafy vegetables by adults. Infants are the most critical element of the population. Aqueous radioiodine can reach man through drinking water and seafood.[1,2]*

The behavior of technetium in the body has been studied, as a result of the use of ^{99m}Tc as a medical tracer. Under such acute dose conditions, technetium administered as the pertechnetate is known to accumulate in the thyroid and/or salivary gland, gastrointestinal tract, stomach, liver, and bladder.[3,4,5,6] Although elimination is quick under acute dose conditions,[3,4,6] few studies examine the consequences of chronic intake of chemical forms that arise from anthropogenic contamination of the environment. From the analysis of 22 human thyroids, Sodd and Jacobs[7] found only 3 which appeared to contain marginally more than the minimum detectable amount, 0.23 pCi (still within the maximum permissible burden for ^{99}Tc in kidney and liver).

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* Superscript numbers designate References appended to this report.

Landa[8] concluded that seedlings of several crops possessed a high uptake of ^{99}Tc with subsequent translocation to aerial tissues. This could provide a potential pathway to grazing animals, but the major pathways to man have not been identified.

A number of references cites the high mobility of ^{99}Tc in groundwater.[9,10] However, slow sorption of ^{99}Tc as a result of organic and/or microbial components of some soils has been demonstrated. Similarly, some soils retain iodine (^{131}I), possibly also as a result of organic and/or microbial soil fractions.[8] Nonetheless, as with ^{99}Tc, radioiodine can be highly mobile in groundwater.[11]

Very sensitive and specific analytical methods for these low specific activity isotopes are required to determine their environmental fate and monitor their release rates. This report describes two such methods: One, for ^{129}I, is a functioning, neutron activation analysis based on the original work of Purkayastha and Martin[12] as refined by Brauer.[13] The other method, for ^{99}Tc, is an isotope dilution technique with mass spectrometric quantification. A unique feature of the analysis concept is the loading of the isolated ^{99}Tc into the mass spectrometer concentrated on a single anion-exchange resin bead. Although the method is still under development, estimates of ^{99}Tc concentrations in a number of environmental aqueous samples have been made by coupling the isolation scheme with liquid scintillation counting.

ANALYTICAL METHODOLOGY

^{129}I

The ^{129}I neutron activation analysis practiced at the Savannah River Laboratory (SRL), an adaptation of Brauer's refinement,[13] uses a simplified radiometric detection system. It has been in use at SRL for over 3 years in a program to define sources and paths of ^{129}I in the surrounding environment in a wide variety of environmental samples. It also simultaneously determines natural iodine, ^{127}I, by its activation to ^{128}I ($t_{1/2}$ = 25 min).

Basically, the analysis consists of four distinct operations: preirradiation isolation of iodine for activation, neutron activation, postirradiation separation and source preparation, and radiometric quantification. These operations are shown in Figure 1.

For aqueous samples, ^{125}I is first added to the 1 L (liter) or more of sample to serve as a yield tracer. The sample is then passed through a 15 × 120 mm, 20-50 mesh, *Dowex** 1-X8 anion exchange column in the Cl^- form. The resin containing the strongly retained iodine is physically loaded into a 480 × 22 mm quartz "inner burn tube" (Figure 2) for pyrolysis/combustion. The inner burn tube is inserted in an all-quartz combustion apparatus and the assembly slowly advanced into a tube furnace at 900°C. A 0.5 L/min nitrogen stream sweeps the pyrolysis gases into a combustion zone supplied with oxygen at 3 L/min, spontaneously forming a stable flame. The

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* *Dowex* is a trademark of Dow Chemical Company.

resultant combustion gases are passed through a 8 × 100 mm column of 8 × 16 mesh activated carbon which quantitatively sorbs iodine species. The flame dies when the pyrolysis is complete, leaving the inner burn tube coated with a carbonaceous residue. By routing oxygen through both the outer and inner burn tubes and slowly withdrawing the apparatus from the furnace, the residue can be burned away leaving only a trace of ash. During the entire process, it is necessary to moderately heat the charcoal absorber to prevent water condensation which lessens the retention of iodine. However, excessive absorber temperatures can expel iodine from the absorber leading to decreased sensitivity through yield loss. A heat lamp, supplemented occasionally by a soft gas flame (hand torch), works well.

Because some volatile organic pyrolysis products can escape complete combustion, especially during sooting episodes (i.e., advancing the sample too rapidly into the furnace), a secondary combustion called a "desmoke" operation is performed (Figure 3). The first absorber, supplied with oxygen, is ignited by a hot flame from a hand torch. As the glowing combustion zone travels down the initial absorber, the gaseous iodine products are trapped in a new charcoal absorber. When combustion of the first absorber is done, a small charcoal absorber is attached to the second absorber and the large charcoal bed is burned in a similar fashion (Figure 4). The final result of this multistep combustion sequence is the isolation and concentration of the sample and spike iodine on a 2.4 x 32 mm bed of charcoal.

The small diameter quartz tube containing the charcoal is cut from the large tube, sealing the small tube's empty end in the process. While under a vacuum, the other end is also sealed closed. Finally, by vigorously heating the charcoal section with an oxygen-propane flame while the empty end is immersed in liquid nitrogen, most of the iodine-containing species are distilled into the empty end of the tube. This is cut and sealed with a hot torch, yielding an evacuated quartz ampoule containing a large portion of the initial sample's iodine content.

Six such ampoules, four containing samples and two containing known amounts of ^{129}I, are sealed in an evacuated aluminum capsule by electron beam or TIG welding. Irradiation is conducted for two hours in a reactor at a neutron flux of about $4 \times 10^{14}/\text{cm}^2$-sec. The pertinent reactions for ^{129}I are:

Analytical Reaction

$^{129}\text{I}\ (n,\gamma)\ ^{130}\text{I} \quad t_{1/2} = 12.5$ hr

Potentially Interfering Reactions

$^{127}\text{I}\ (n,\gamma)\ ^{128}\text{I} \quad t_{1/2} = 25$ min

$^{127}\text{I}\ (n,2n)\ ^{126}\text{I} \quad t_{1/2} = 13.1$ days

$^{127}\text{I}\ (n,\gamma)\ ^{128}\text{I}\ (n,\gamma)\ ^{129}\text{I}\ (n,\gamma)\ ^{130}\text{I}$

$^{125}\text{I}\ (n,\gamma)\ ^{126}\text{I}$

Following irradiation, the ampoules are removed from the capsule underwater and carefully cleaned. Their contents are subjected to a postirradiation separation consisting of redox-controlled distillation and solvent extraction steps designed primarily to minimize ^{82}Br contamination.

In the first stage, bromide and iodide carriers are added to the contents of the ampoule. After oxidation by permanganate, bromine is distilled from solution leaving iodate in the heels. Reduction of the iodate by metabisulfite is followed by more bromide carrier addition, hydrogen peroxide oxidation, acidic hydroxylamine reduction, and distillation of the resultant elemental iodine into a metabisulfite receiving solution.

Analogously, redox-controlled extraction of I_2 into xylene is conducted giving a final solution containing the radioactive iodine and its carrier. These are precipitated as silver iodide and collected on a 25-mm-diameter membrane filter. The counting source is finished by mounting the filter between two pieces of masking tape.

After a suitable time for ^{128}I decay (i.e., 13 hr), ^{130}I activity is quantified at 536 keV with a 44 × 71 mm Ge(Li) (coaxial lithium-drifted germanium) detector (Ortec Model 8001-1523) and a Canberra Model 8100 multichannel analyzer. Data recording and reduction are performed by a Hewlett-Packard Model 9830B calculator with printer. A few days later, when ^{130}I activity has decayed, ^{125}I activity is determined for the samples, comparison standards, and an ^{125}I reference source. (The latter source is quantitatively prepared from the amount of ^{125}I added to each sample and standard. It serves as a check on the yield of the isolation scheme but is not used to calculate the ^{129}I results.) A 1000 mm^2 LEP (low energy photon) detector (Princeton Gamma-Tech Model IG 1012) is used.

If the amount of ^{129}I added to the comparison standards and the ratios of the decay-corrected ^{130}I activity to the ^{125}I activity for the samples and comparison standards are known, the ^{129}I content of the samples may be calculated.

Under the above conditions and Currie's definition[14] of the quantification limit, L_q, as little as 3.8 fCi of ^{129}I can be determined (s_{rel} = 10%) with a 30-minute counting period. Longer counting periods can increase the sensitivity. Brauer et al.[13] and Keisch et al.[15] discuss a wide range of detection systems suitable for ^{129}I analysis.

Typical analysis performances based on a full-time analyst and a part-time counting technician are:

- an isolation yield of about 50%,
- irradiation of 2 capsules per week (8 samples) on the average, and
- approximately 8 days from submission of the sample until results are in hand.

^{99}*Tc*

As indicated by Kaye et al., an isotope dilution technique utilizing a surface ionization mass spectrometer yields the lowest sensitivity limit yet attained for ^{99}Tc.[16] Work is in progress at the Savannah River Laboratory to develop a chemical procedure to isolate ^{99}Tc from environmental samples for mass spectrometric analysis. A 3-stage mass spectrometer with high abundance sensitivity is currently under construction. The mass spectrometry group at Oak Ridge National Laboratory has provided contract interim mass spectrometric support for the development work. The concept, progress, and results from aqueous environmental samples are described.

The isotope dilution concept is based on measurement of the ratio of ^{99}Tc and ^{97}Tc in the material obtained from the isolation scheme. A known amount of ^{97}Tc is added to the sample to serve as a yield tracer. Since isobars at masses 97 and 99 can interfere with the determination of the technetium mass ratio, the isolation procedure must be designed to strongly discriminate against elements other than technetium at these masses. The most serious potential interference is ^{97}Mo, with ^{99}Ru of lesser concern.[16] Some correction can be applied for ^{97}Mo by observing ^{98}Mo, but the amount of correction is limited by statistical considerations of the measurements.

The concept and current state of the isolation scheme are shown in Figure 5. In order, the aqueous sample is subjected to: cation-exchange chromatography, anion-exchange chromatography, evaporation, two stages of solvent extraction, and concentration of its technetium onto a single anion-exchange resin bead for mass spectrometry. If the ^{99}Tc concentration is high enough in the sample, liquid scintillation counting may be substituted for the last operation.

Technetium is expected to exist in an anionic state in the initial sample, and hydrogen peroxide is added to assure TcO_4^-. The initial strong acid cation-exchange step thus retains no technetium while providing decontamination from cationic species. On the other hand, the subsequent strong base anion-exchange step retains technetium, molybdenum, and ruthenium among others. The first discrimination against the latter two elements is attained in this step by utilizing the fact that 1M hydrochloric acid elutes molybdenum and ruthenium but not technetium. Technetium is easily eluted with 4M nitric acid.[17] Based on a 2-L sample size, a 150-mL elution volume, and a typical 88% yield of technetium, a nearly 12-fold concentration is achieved.

Evaporation of the eluate follows and accomplishes three things: It reduces the volume of the eluate to a value that is compatible with the scale of the subsequent solvent extraction steps. It further concentrates technetium. And, it removes most of the nitrate introduced by elution with nitric acid during anion-exchange chromatography. The last function is particularly important since nitrate will depress the technetium distribution coefficient in the first solvent extraction. Typically, a final evaporation volume of 0.5 mL is sought since this reduces the amount of nitrate sufficiently. The technetium loss during evaporation to date has ranged from none to nearly 50%. This step can constitute the largest yield loss in the procedure and requires further development. Such behavior is well known in the literature.[18]

Two identical stages of solvent extraction follow. Tetrapropylammonium pertechnetate is extracted into methyl isobutyl ketone in an ion-association solvent extraction patterned after the work of Maeck et al.[19] Back-extraction into an aqueous phase is accomplished by diluting the methyl isobutyl ketone with cyclohexane.[20] A single stage of extraction/back-extraction gives nearly quantitative recovery of technetium at moderate nitrate concentrations while providing a decontamination factor of at least 8 x 10^4 for molybdenum. In practice, at a nitrate concentration of 0.9M, 95% of the ^{99}Tc in tracer solutions is routinely recovered by the two-stage solvent extraction.

For very low levels of ^{99}Tc, loading onto a single anion-exchange bead would follow. In development studies, the aqueous back-extract from the solvent extraction steps was evaporated to 0.1 mL in a conical, screw-top, 15 mL centrifuge tube containing a single anion-exchange bead ($\sim$0.1-mm diameter, AG 1X8, Cl^-). After 2 hours contact time with vigorous agitation, equilibrium ^{99}Tc uptakes of 75 to 80% were obtained. Amounts as low as 0.03 ng of ^{99}Tc were loaded.

A number of these resin beads has been submitted for mass spectrometric analysis to R. L. Walker at Oak Ridge National Laboratory. Each bead was mounted directly on a "V" filament of a surface ionization source mass spectrometer for analysis. The results indicate an average 50-fold increase in ^{99}Tc ionization efficiency (relative to evaporative application of the same amount of ^{99}Tc in solution) over the range of 0.03 to 30 ng. Localization of the ion source by this technique should provide a proportionate increase in the sensitivity of the method compared to solution loading. Resin bead handling techniques have been worked out during development and pose no obstacle to the practical use of the concept.

If the initial sample contains ≥10 pCi of ^{99}Tc, liquid scintillation detection can be conveniently used to quantify the results. The 0.15-MeV ^{99}Tc β^- can be efficiently counted under the same conditions used for ^{14}C. The presence of ^{99}Tc in such samples can be verified by comparing their β-ray absorption curves in aluminum with the curve from an authentic ^{99}Tc source. Mass spectrometry can also be used to establish the presence of a mass-99 species.

None of the results has yet been generated by an actual isotope dilution procedure due to the lack of ^{97}Tc yield tracer. Rather, all environmental values have been calculated by assuming that the technetium yields obtained with the samples were the same as those from standards run through the same procedure. Because underestimates of ^{99}Tc can result from lower than expected recovery, the environmental values are probably lower limits.

^{97}Tc can be prepared by neutron irradiation of ^{96}Ru. The detailed procedure has been described by J. H. Kaye.[16] Preparation of ^{97}Tc from enriched ^{96}Ru is desirable because of high yield and minimal ^{103}Ru formation. The latter simplifies the handling during postirradiation isolation. However, due to uncertain availability of ^{96}Ru, irradiation of natural ruthenium is also being investigated at the Savannah River Laboratory. Isotope dilution with ^{97}Tc will allow accurate yield determination and permit the use of resin bead mass spectrometry as a quantitative technique.

AQUEOUS TRANSPORT MEASUREMENTS

The scope of the environmental analysis differs depending on the isotope involved. Because the ^{129}I method is well-developed and of high sensitivity, a greater range of samples can be analyzed than with the developmental ^{99}Tc procedure. Samples analyzed for ^{129}I to date are from seepage basins receiving low level radioactive wastes from fuel reprocessing areas; monitoring wells between the seepage basins and a nearby stream; a surface spring receiving seepage from the basins and draining into the stream; a number of surface streams traversing or originating in the Savannah River Plant (SRP) site; and the Savannah River above and below SRP. The sensitivity of the current ^{99}Tc analysis has limited its application to seepage basin and spring samples.

Table 1 summarizes both the ^{129}I and ^{99}Tc results for the two different seepage basin systems and spring samples. The soils at this site consist of sandy clay, clayey sand, silt, and clay. Nine groundwater monitoring well samples also analyzed for ^{129}I ranged from 0.083 to 297 pCi/L. The highest values correlated well with known major groundwater movement patterns determined earlier from ^{3}H migration studies. From these results, it can be concluded that both ^{129}I and ^{99}Tc move freely from the seepage basins with the groundwater.

Analyses of the stream receiving the seepage from the basin system, other on-plant streams, and the Savannah River are presented in Table 2. Four Mile Creek conveys most of the aqueous ^{129}I from SRP to the off-plant aqueous environment. This is expected since Four Mile Creek receives the seepage from the two major seepage basin systems. Its $^{129}I/^{127}I$ ratio was found to be 17×10^{-6} , over 14 times that of any other SRP stream. A threefold increase in the ^{129}I concentration of the Savannah River is noted as it flows past SRP and receives the water from Four Mile Creek and other streams.

Based on known flows at the time these samples were taken, the increase in ^{129}I concentration measured in the Savannah River is equivalent to an output rate of 27 mCi/yr. On the other hand, based on Four Mile Creek measurements and flows, the ^{129}I output rate is estimated to be only 16 mCi/yr. The difference appears to be outside the bounds expected from analytical imprecision and awaits additional sampling and analysis for explanation. Routine samples of the Savannah River up- and downstream of SRP are now being taken to establish a larger data base and a more confident estimate of annual aqueous emission. In all cases, the concentrations found in the Savannah River have been several orders of magnitude below the Federal concentration guides for releases to uncontrolled areas.[21]

CONCLUSIONS

These results demonstrate that both ^{129}I and ^{99}Tc from seepage basin systems move easily through the sandy clay, clayey sand, silt, and clay soils at the Savannah River Plant to the external aqueous environment. Coupled with demonstrations of mobility at the Hanford Reservation, this work points out the need for monitoring the aqueous environment around potential sources of ^{129}I and ^{99}Tc such as waste storage tanks and low level aqueous waste disposal systems supporting fuel reprocessing operations. The $^{129}I/^{127}I$ analysis provides established methodology of very high sensitivity for this task. ^{99}Tc can now be estimated in aqueous samples of reasonable size containing more than 10 pCi. Further development of this methodology should ultimately provide detection in the 0.2 to 2.0 fCi range.

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TABLE 1

^{129}I and ^{99}Tc in Seepage Basin and Spring Samples

Sample	^{129}I, *pCi/L*	$^{129}I/^{127}I$ *Atom Ratio*	^{99}Tc, *pCi/L*
Seepage Basin F-3	140 ±4	9.9×10^{-3}	700[a]
Seepage Basin H-4	74 ±3	7.2×10^{-3}	90 106
Spring	224 ±6	-	291
Spring	-	-	204[a]
Spring	-	-	183

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[a] Presence of ^{99}Tc in solution isolated by procedure verified by mass spectrometry and β^- half-thickness measurement.

TABLE 2

^{129}I in Stream and River Samples
(All samples taken 8/25/77)

Sample Location	^{129}I, *fCi/L*	$^{129}I/^{127}I$ *Atom Ratio*
Four Mile Creek[a]	43 ±2	17 × 10^{-6}
Upper Three Runs[b]	1.1 ±0.3	1.2 × 10^{-6}
Lower Three Runs	0.2 ±0.2	0.1 × 10^{-6}
Steel Creek	1.3 ±0.2	0.5 × 10^{-6}
Savannah River (Upstream)	1.6 ±0.5	0.5 × 10^{-6}
Savannah River (Downstream)	5.2 ±0.4 5.4 ±0.3 (duplicate)	1.6 × 10^{-6}

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a. Major stream receiving seepage from basin system.

b. Originates off-plant. Other creeks originate on-plant.

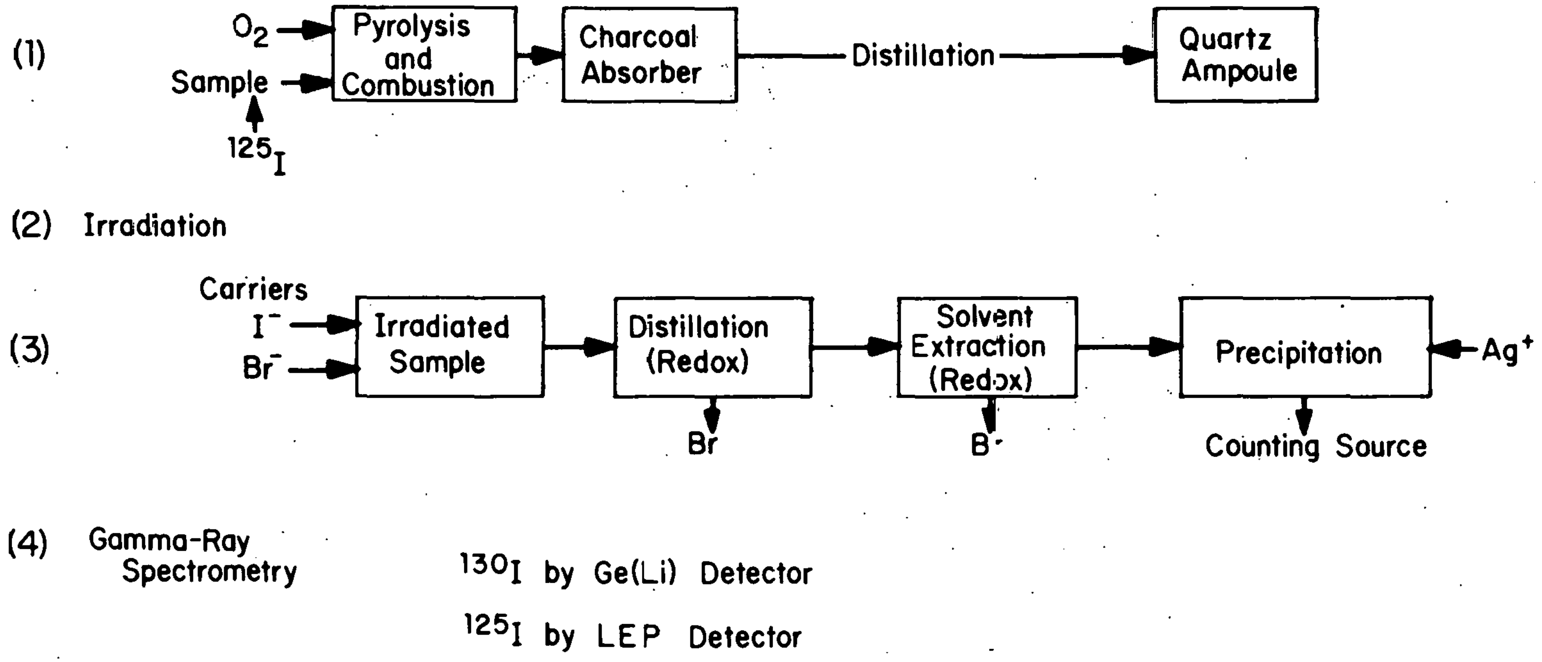


FIGURE 1. ^{129}I Analysis Scheme

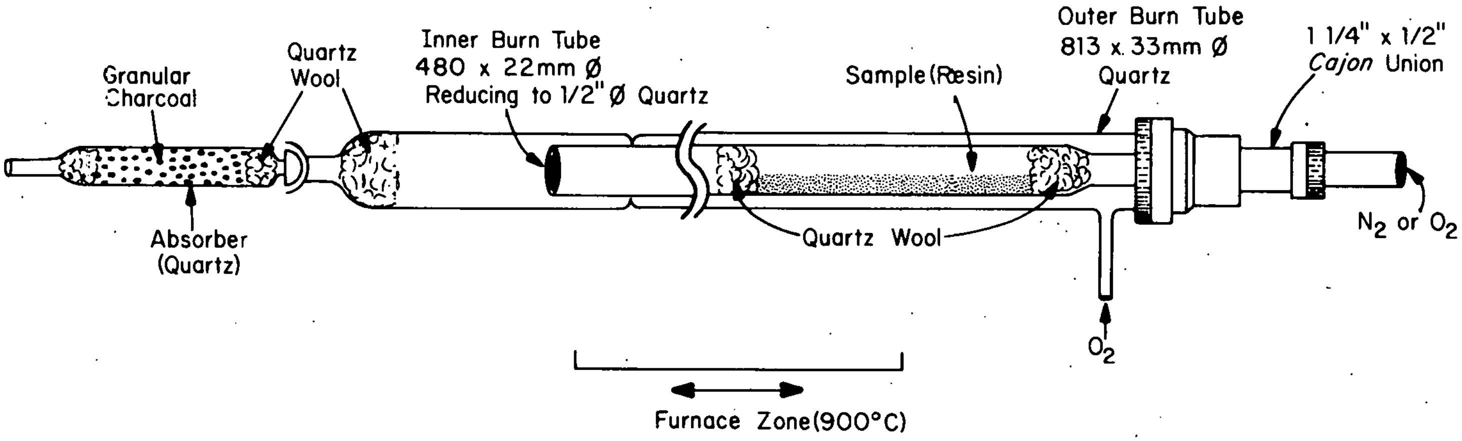


FIGURE 2. Combustion Apparatus

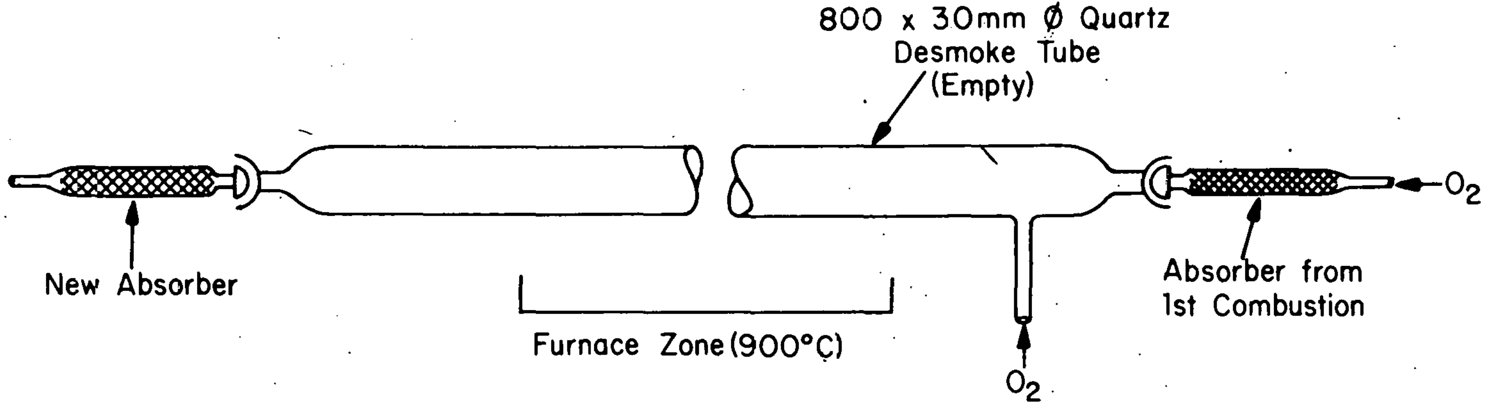
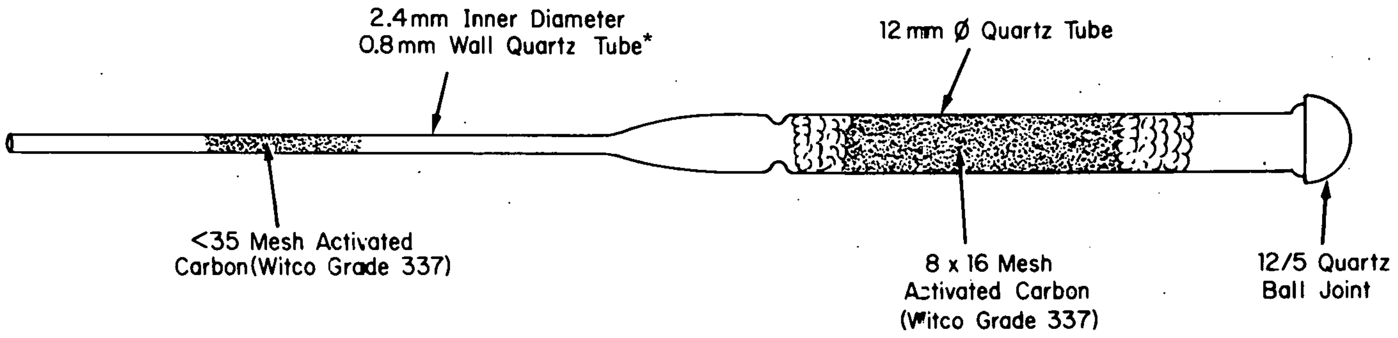


FIGURE 3. Desmoke Apparatus



* Attached just prior to second absorber combustion.

FIGURE 4. Final Absorber

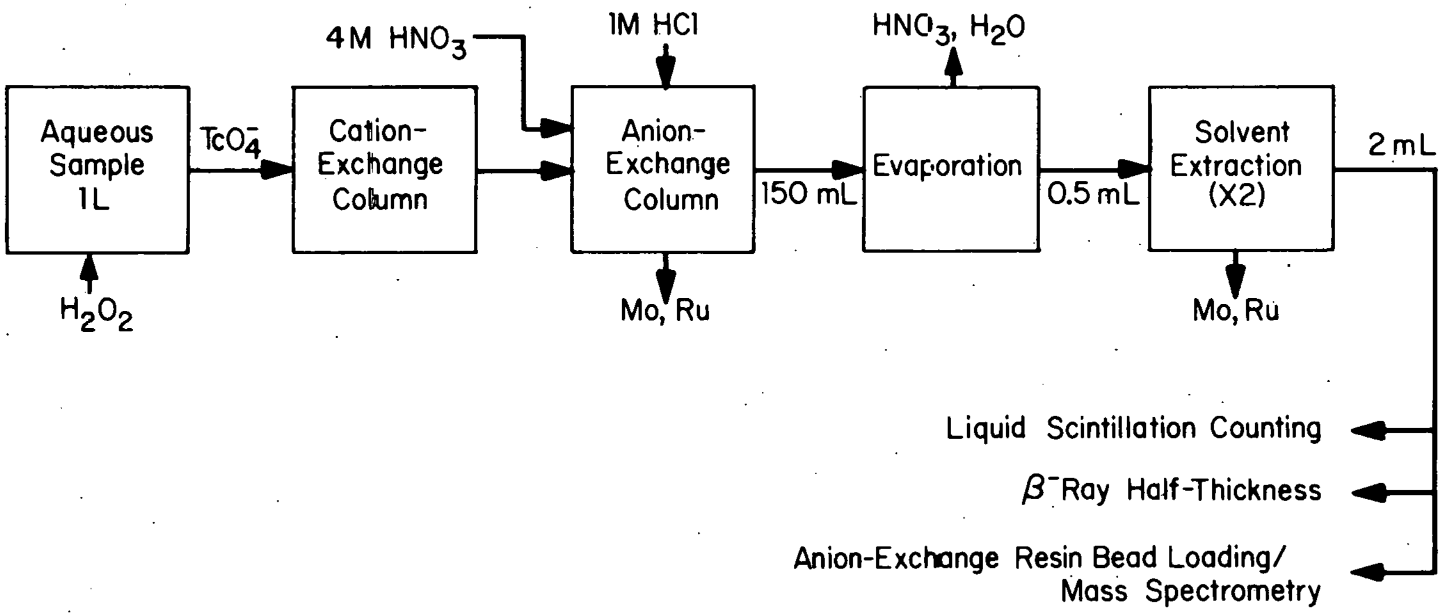


FIGURE 5. Technetium Analysis Scheme