Radioactive Waste Disposal

Deiodination and Solidification of Radioassay Waste

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We describe our efforts to produce a successful method to purify the liquid I-125 waste generated from radioassays. The resulting liquid is equivalent to background and can be legally disposed of in public sewage without dilution, after the radioactivity is transferred to a small volume of solid waste. In our procedure liquid waste is acidified with 70% perchloric acid, filtered, then passed through anion exchange columns. With 60 to 90% of the original activity removed, the remainder is soaked on a bed of activated charcoal. After 30 min the remaining activity is below legal limits for disposal for I-125 (88.8 dpm or 4 × 10^{-5} μCi/ml). After soaking overnight in charcoal the liquid is equivalent to background levels. This method requires no more time or expertise than a routine radioassay does. In addition, the reagents that we use for this procedure last for about ten weeks, thus reducing our total volume of liquid radioactive waste by almost 12-fold.

There are only three commercial disposal facilities for low-level radioactive waste in the United States. Regulations governing low-level waste come from numerous federal and state agencies. The Department of Transportation regulates shipping, the Nuclear Regulatory Commission regulates the type and amount of radioactivity, and further regulations for each disposal site are imposed by the individual states involved. The limited number of sites, their long distances from many users, the complex regulations regarding packaging and shipping, and the possibility of future shutdowns of all sites (such as occured in October to November 1979) (*1*) make disposal of radioactive waste a costly and problematic endeavor for any institution. Although medicine generates a small portion of the total of all nuclear wastes, reduction of the volume of medically-generated radioactive waste might be a partial solution to the problem (*2*).

Iodine-125, with a half life of 60 days, is the longest lived medical radioisotope that leads to a significant volume of liquid radioactive waste and is generated primarily from radioassays. Although in California, I-125 in liquid can be disposed of in public sewage if diluted to 4 × 10^{-5} μCi/ml or 88.8 dpm/ml (*3*), many institutions solidify it and ship it to one of the disposal sites. We attempted to convert liquid radioassay waste containing I-125 to a small volume of solid and reduce the level of I-125 in the remaining liquid to background levels for disposal in public sewage without dilution.

Materials and Methods

One liter per week of liquid waste containing I-125 is accumulated by our laboratory in the process of assaying 1,000 patient samples per month. The waste is from various radioassays including T_3RU, T_4, T_3 RIA, free T_4, ferritin, CEA, gastrin, renin, and HCG-β. All experiments were conducted under a fume hood.

Acidification and Anion Exchange: We designed an experiment to test the effectiveness of two separation and solidification methods—anion exchange resin at varying resin concentrations and protein denaturation with perchloric acid at differing pH levels. Precipitates in acidified waste were removed by filtration through glass wool prior to anion exchange of supernatant. Various pH levels were tested by preparing three 200-ml aliquots; the first was left at pH 5.9, while the second and third were acidified to pH 4.5 and 2.5, respectively. The pH 5.9 and 4.5 aliquots were equally divided and slurried with 10, 20, and 30 g of AG1X-2 (Cl^-, 100–200 mesh) resin (Bio-Rad Laboratories) in beakers. The pH 2.5 aliquot was processed through 10, 20, and 30 g of the same resin in columns. The columns were 60-cc syringe barrels plugged with glass wool and fitted with a 3-way stopcock. One-ml aliquots of supernatant or effluent were removed at hourly time intervals and counted for I-125.

To determine optimal resin concentration, another 200-ml aliquot of waste was acidified to pH 2.5 with perchloric acid and precipitates were removed by glass wool filtration. The supernatant was placed in a beaker with a magnetic stirrer. Every 5 min a 1-ml aliquot was

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removed for counting and resin was added in 10-g aliquots until a concentration of 0.5 g of resin per ml of waste was reached.

Another anion exchange resin, AG1X-8 (acetate, 100–200 mesh), was tested to determine if it was more effective that AG1X-2. Because of the larger cross-linking of AG1X-8 resin we thought that it might make the acid denaturation step unnecessary, and also remove CEA, a large radioiodinated glycoprotein that was present in all the previous waste aliquots and was not precipitated by acid. Three 200-ml aliquots of waste were prepared. The first contained waste from all RIA procedures including CEA and was not acidified. The second contained no CEA waste and was also not acidified. The third was identical to the second but was acidified with 70% perchloric acid to a pH of 2.5. Each aliquot was halved and processed through AG1X-2 and AG1X-8 resin columns. Each column contained 35 g of resin, with a flow rate of 6 ml/min. One-ml aliquots were collected for counting with no time allowed for incubation.

Activated Charcoal: Two charcoal adsorption techniques for further purification were tested. One method used static incubation of 30-ml resin-processed waste in a column containing 20-g charcoal (Aquarium Pharmaceuticals). The other method used incubation of 200-ml aliquots with 30-g charcoal in a beaker with continuous mixing by a magnetic stirring bar. One-ml aliquots were removed for counting at various time intervals from 5 min to 24 hr. To test for saturation, the charcoal that had been mixed was filtered, then incubated with a fresh 200-ml aliquot of resin-processed waste. One-ml aliquots were removed for counting at 30, 60, and 90 min.

Summary of Protocol: After testing a variety of methods including variations of those described above, we chose the following protocol (Fig. 1) for routine use:

1. Adjust pH of waste to 2.5 with 70% perchloric acid, mix for 15 min with a magnetic stirring bar, and let precipitates settle for at least 15 min.
2. Filter through glass wool into a 1000-ml reservoir above a 60-ml column containing 35–40 g of AG1X-8 (acetate form; 100–200 mesh) anion exchange resin. Adjust stopcocks of reservoir and anion exchange column to a flow rate of 6 ml/min.
3. Let the effluent from the anion exchange column drain into a closed 1000-ml reservoir containing 150 g of activated charcoal.
4. After at least 1-hr incubation, drain the liquid from the charcoal reservoir. Remove 1-ml sample to determine the I-125 content.

When the activity level is below 88.8 dpm/ml the liquid can be disposed of in sewage without any dilution problems. We have used this method to purify approximately 1 liter of liquid I-125 waste every week for ten weeks using the same reagents.

Results

Anion Exchange: In the initial experiments (Table 1, A) the resin removed about 90% of I-125 from liquid waste in 2 hr or less without acid precipitation, leaving most of the radioactivity in a small volume of solid. More than 96% of the I-125 was removed from the liquid when acid denaturation was combined with anion exchange (Table 1, B). Although over 98.5% of the radioactivity could be converted to a solid, the liquid and solid phases were still in one beaker and had to be separated manually. Anion exchange in columns proved

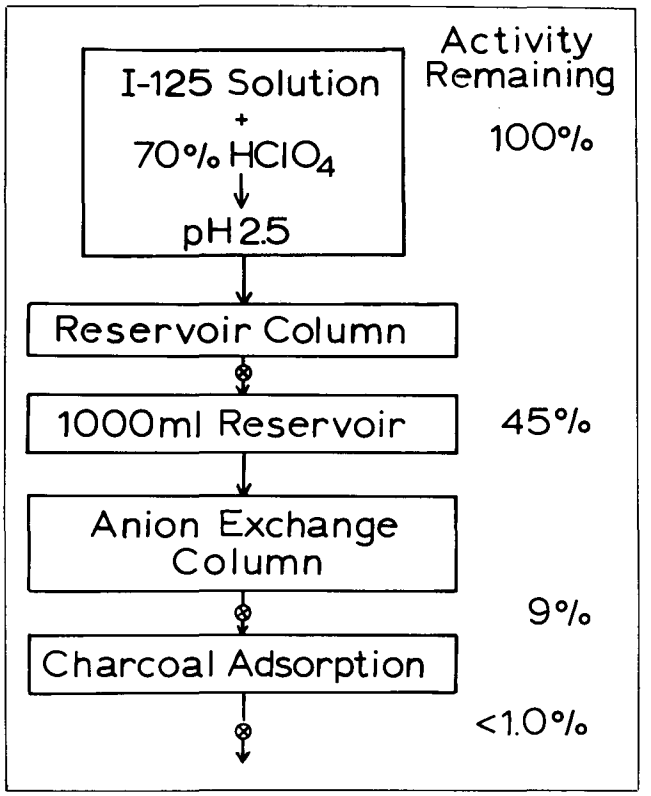

FIG. 1. Denatured proteins are separated from liquid by glass wool in a 60-cc syringe barrel, which drains into a 1-l reservoir with a 60-cc syringe attached for anion exchange. The stopcock controls flow to 6 ml/min and drains into a bed of activated charcoal with glass wool for filtration.

TABLE 1. Percent of I-125 Remaining in Liquid after Acidification and Anion Exchange

			% I-125 remaining after incubation with resin			
Method	pH	Gram resin/ 200-ml waste	1 hr	2 hr	4 hr	24 hr
A) Beaker	5.9	10	—	11.1	11.0	10.4
		20	—	7.7	7.6	8.0
		30	—	4.3	4.4	4.1
B) Beaker	4.5	10	—	3.9	4.0	4.0
		20	—	3.5	3.5	1.6
		30	—	1.5	1.5	1.6
C) Column	2.5	10	13.2	13.1	13.0	15.4
		20	12.4	12.6	16.1	16.1
		30	9.2	9.5	10.5	12.3

practical for separating liquid and solid phases but was somewhat less efficient for removing radioiodine (Table 1,C).

In the experiment designed to determine optimal resin concentration, the amount of I-125 in acid-denatured waste steadily decreased with increasing concentrations of resin until a concentration of 0.25 g/ml of waste was reached (Fig. 2). Although 90% of the original I-125 was removed by perchloric acid denaturation, filtration, and AG1X-2 (Cl^-) resin, the amount of radioactivity remaining in the liquid was still about 30 times higher than legal for sewage disposal without dilution.

The AG1X-2 and AG1X-8 resins were equally effective. With CEA present in the waste and no acid denaturation, the AG1X-8 resin removed 83% of the radioactivity; the AG1X-2 removed 82%. Combined with acid denaturation and filtration, a total of 92% of the radioactivity was removed using AG1X-8 and 91% was removed using AG1X-2. In the absence of CEA and without acid denaturation both resins removed about 65% of the radioactivity; with acid denaturation they removed 86%. Both resins functioned equally well regardless of the presence or absence of CEA molecules.

Charcoal: The charcoal adsorbed additional radioactivity from the resin-processed waste. After 30 min of soaking in 0.67 g of charcoal per ml of waste, the legal level for disposal without dilution was reached. After soaking overnight, the activity remaining in the liquid was equivalent to background levels (Table 2,A). Using the mixing method, 0.15 g of charcoal per ml of waste reduced the activity to background levels in 60 min (Table 2,B). When the charcoal was filtered and reused on a fresh aliquot of waste, more radioactivity was adsorbed (Table 2,C). The two adsorption methods resulted in a trade-off. Columns required more charcoal but conveniently separated liquid from solid. The charcoal mixing method pulverized the charcoal, thus requiring centrifugation and filtration before counting and disposing of liquid.

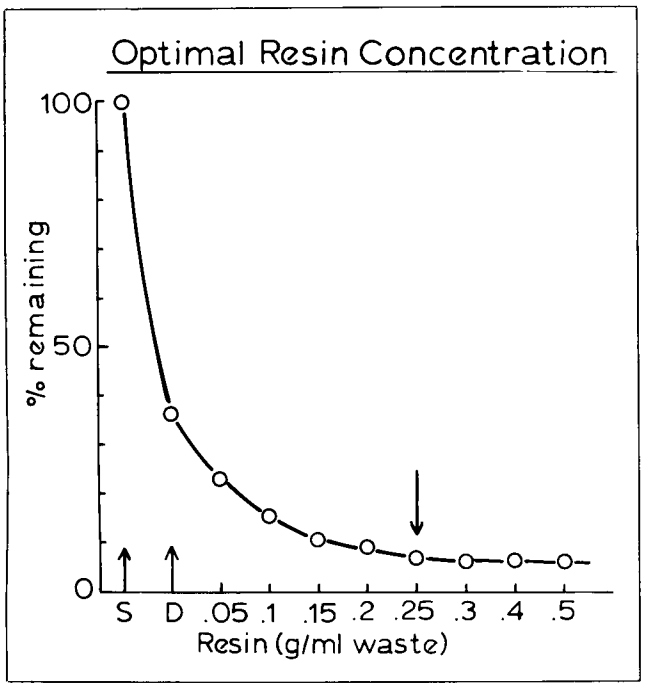

FIG. 2. Starting with 3.2 μCi, 40% remained in solution after acid denaturation. As resin concentration increased, amount of I-125 remaining in solution decreased until a concentration of 0.25-g resin per ml of waste was reached.

Discussion

Used weekly for ten weeks, 150 ml of 70% perchloric acid, 35 g of AG1X-8 resin, and 150 g of charcoal have been used to purify a total of 8.4 l (2.2 gallons) of radioactive waste containing about 163 μCi of I-125. The amount of radioactivity remaining in the liquid after purification was equivalent to background levels. The

TABLE 2. Amount of I-125 Remaining in Liquid after Incubating Resin-Processed Waste in Activated Charcoal

Method (charcoal concentration)	Incubation time	dpm/ml remaining	% remaining
A) Column	initial	14,088	100.0
0.67 g/ml	after acid/resin	1,282	4.1
	5 min	170	1.2
	30 min	30	0.2
	60 min	30	0.2
	24 hr	<2	<0.01
B) Mixing	initial	17,416	100.0
0.15 g/ml	after acid/resin	2,386	13.7
	30 min	332	1.9
	60 min	26	0.1
C) Mixing	initial	28,714	100.0
0.15 g/ml	after acid/resin	2,010	7.0
(same charcoal	30 min	835	3.1
as B with fresh	60 min	533	2.2
waste)	90 min	55	0.2

TABLE 3. Costs of Disposing of I-125 Waste by Solidification with Cement and Shipment to Burial Site Versus Purification and Conversion Method

Item	Cement solidification $/28-32 gallons	Conversion method $/gallon
55-gallon, epoxy-lined drum	17	—
Cement	11	—
Sand	12	—
Personnel time	25	0.50
Acid	—	2.00
Anion exchange resin	—	1.50
Charcoal	—	0.60
Shipping cost	110.00	negligible
Cost/gallon	$5.83	$4.60
Volume change	50% increase	93% decrease

radioactivity originally contained in the liquid was converted to a small volume of solid—resulting in a 93% reduction in the volume of waste. Some I-125 began eluting from the anion exchange resin after being used for ten weeks.

Although other methods were tested, the method we have described requires a minimum of time and expertise and has been used routinely on all our waste without upsetting the laboratory's assay schedule.

The estimated costs of disposing of our liquid waste by solidifying and then shipping to a burial site and the costs of conversion and sewage disposal are given in Table 3. Cement solidification is accomplished by mixing 180 lb of highly adsorbent "floor dry" sand with 140 lb of cement and 30 ± 2 gallons of radioactive waste. The amount of radioactive liquid the cement is able to adsorb depends on the adsorbency of the sand, humidity, and temperature. The cement mixture is poured into 55-gallon drums to solidify overnight. Four 55-gallon (520-lb) drums are shipped weekly from our university to Nevada for burial. Personnel costs were estimated but capital equipment costs (cement mixers, shovels, storage facilities, etc.) were not included. It should be noted that the volume of solid remaining is smaller and lighter than commonly used adsorbing materials. This method results in at least a 12-fold volume reduction while cement adsorption increases final volume by 50%. The largest fraction of solid waste remaining is charcoal, which can be compacted to reduce its volume prior to packaging for disposal.

References

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