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CONTAINMENT OF IODINE-131 RELEASED BY THE RALA PROCESS

G. K. Cederberg
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October 30, 1961

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**NATIONAL REACTOR TESTING STATION
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A B S T R A C T

Uncontrolled releases of large amounts of iodine occurred during the early stages of RaLa operation at the Idaho Chemical Processing Plant. A ten-fold reduction in the iodine content of the off-gas was achieved by process modifications, primarily the addition of mercury salts to the acidic process solutions. An additional ten-fold reduction was obtained by installing an activated charcoal adsorption unit in series with the original iodine removal scrubber. The iodine content of particulate entrainment limited the overall iodine removal efficiency of the revised RaLa off-gas iodine removal system.

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SUMMARY

Radioactive iodine concentrations in the off-gas from the RaLa (barium-140 recovery) process during the initial runs were considerably in excess of design levels. A dual approach was made to the mitigation of this problem. First, the operating procedures, equipment designs, and chemistry of the process solutions were reviewed and modified as necessary to improve the performance of the original system. Second, a new charcoal bed adsorption unit was designed and installed to further reduce the quantity of iodine carried by the off-gas.

The addition of mercury salts to several of the process solutions was credited with the major part of the improvement of iodine removal resulting from modifications to the original system. The off-gas scrubber solution was changed from one molar sodium hydroxide to 5 weight per cent nitric acid, with 0.001 molar mercuric nitrate and 0.001 molar mercurous nitrate added. Similar concentrations of mercury salts were added to the other acidic solutions used in the process. The change in process solution composition, plus some minor equipment and procedure modifications, lowered the iodine concentration in the RaLa process off-gas by a factor of about ten.

The factor limiting the attainable iodine removal appeared to be the presence of dusts or mists containing iodine compounds. Consequently, the filtration efficiency of an iodine removal unit is as an important a design feature as is the gaseous iodine removal efficiency.

I. INTRODUCTION

A problem at the Idaho Chemical Processing Plant (ICPP) has been the containment of radioactive iodine during operation of the RaLa process. This process is used to recover barium-140 from a two-day cooled MTR fuel element (1,9). The very short cooling time (fuel elements are normally cooled at least one hundred days before processing) results in a fission product content much higher than is usually encountered. Iodine-131, half-life of 8.05 days, has been the most troublesome of the short half-life fission products characteristically encountered in RaLa operations. Small, but significant, quantities of the iodine are evolved during several of the process steps, and the required containment measures have proved unexpectedly complex.

The RaLa process includes dissolution of the fuel element, separation and purification of the barium-140, isolation of the uranium for subsequent recovery, and disposal of the process wastes. The uranium-aluminum alloy MTR element is dissolved in 8 molar sodium hydroxide, forming a precipitate of uranium and fission products and a supernatant sodium hydroxide-sodium aluminate solution. About 10 per cent of the iodine is found in the precipitate and the remainder in the supernate, with essentially no volatilization of iodine at this point. The precipitate is separated in a batch centrifuge; the supernate is discarded to waste storage. The subsequent process steps to separate and purify the barium-140 are performed in the bowl of this centrifuge. These steps, involving the repeated use of a strong oxidant (concentrated nitric acid), generate most of the gaseous iodine found in the RaLa process off-gas system. Most of the iodine found in the vessel off-gas system is released during solution transfers, particularly transfer of the process wastes to storage. The use of compressed air to inject reagent solution into the centrifuge bowl, and the consequent escape of iodine-contaminated gases from the centrifuge shaft seal, is the major cause of iodine release to the cell atmosphere and ventilation system.

Each of the three iodine contaminated systems--process off-gas, vessel off-gas and cell ventilation air--is capable of releasing hazardous quantities of radioactive iodine to the atmosphere. The iodine containment measures originally provided were adequate only for the process off-gas, and then only during favorable weather (atmospheric diffusion) conditions. This weather dependency and the possibility of iodine contamination of the vessel off-gas or cell ventilation air seriously reduced the overall factor of safety for the RaLa process. The possible consequences of an excessive release of iodine to the inhabited parts of the ICPP, or to the atmosphere during poor diffusion conditions, were sufficiently severe to justify an intensive program to improve the iodine containment measures.

II. ORIGINAL IODINE REMOVAL SYSTEM

The virtually complete containment of iodine was a major objective in the design of the RaLa process. The RaLa system was designed for a vacuum of 10 to 20 inches of water in all process equipment, to insure that any leakage would be from the cell atmosphere into the equipment. All the process off-gas, plus any air leakage into the system, was to be passed through an iodine removal scrubber before release to the atmosphere through the 250-foot plant stack. The scrubber--a column 8 inches in diameter, by 7 feet long, packed with 1/2-inch long sections of 3/4-inch, Schedule 40, stainless steel pipe--was designed to lower the iodine content of the off-gas to a level safe for disposal. The one molar sodium hydroxide scrubber solution was recirculated at the rate of 8 gpm with a nitrogen lift.

The first processing of a fully irradiated fuel element disclosed serious deficiencies in the iodine containment measures. The peak iodine release rate to the process off-gas, which occurred during dissolution of the uranium-fission product precipitate, was considerably higher than had been anticipated. The maximum iodine content of the scrubbed off-gas was sufficiently low to permit operation during the most favorable atmospheric conditions, but it was a factor of 100 too high to allow operation of the RaLa process during poor diffusion conditions.

The transfer of the dissolution supernate waste solution, through the plant lines and vessels to permanent storage, released about 200 curies of iodine-131 to the main ICPP vessel off-gas system. This solution of sodium hydroxide and sodium aluminate was neutralized with nitric acid prior to transfer to prevent the precipitation of hydrous aluminum oxide. About 90 per cent of the iodine originally contained in the fuel element was present in this solution, and a significant part of this iodine was readily liberated from the acidified solution. The transfer of the waste could be deferred until weather conditions were favorable, but the quantity of iodine released was still a serious hazard.

The centrifuge vent could not handle the sudden burst of air that accompanied the injection of solutions into the centrifuge. Consequently, the centrifuge pressurized and iodine-contaminated gas was forced through the shaft seal into the cell atmosphere. The shaft seal did perform properly during steady state operation.

Pressure surges occasionally blew the liquid from the seal in the line connecting the RaLa off-gas line to the ICPP liquid waste collection line. Loss of the seal liquid allowed the escape of RaLa off-gas into other ICPP equipment through the liquid waste collection system.

III. SYSTEM MODIFICATIONS

The release of iodine in excessive quantities and at undesirable locations made remedial action imperative. The first step taken was the review and improvement of operating procedures. The second step was the modification of any malfunctioning equipment. The third step was a change in the chemical composition of some of the process solutions. The steps were generally taken in the order listed, but there was a considerable chronological overlap. Quantitative measurements of the effects of each change were not attempted, because the unique iodine content of each fuel element would have obscured the results.

A. Procedure Changes

The pressure of the nitrogen and air used in the jet syphons for solution transfer was lowered. Iodine was being volatilized from acidic solutions and entrained from basic solutions during transfers. The reduction in pressure of the motive media reduced the iodine release. The frequency and duration of solution sparging and the pressure of the sparge air were reduced for similar reasons and with similar results.

The temperatures of solutions being sparged were lowered in a number of instances in a successful effort to reduce the volatility of the dissolved iodine.

The procedure for injection of solutions into the centrifuge bowl was revised to reduce the quantity of compressed air used to move the solutions. The elimination of the large excess air previously used helped prevent pressurization of the centrifuge and escape of iodine through the shaft seal to the cell atmosphere.

B. Equipment Changes

An improved centrifuge shaft seal was designed and installed. The new seal incorporated an air purge of the shaft opening to keep the iodine-containing off-gas in the housing of the centrifuge.

The liquid seal depth in the RaLa off-gas drain line was increased from 3 feet to 6 feet, so that minor pressure fluctuations would not blow the seal and allow off-gas to escape into the main ICPP process equipment waste system.

C. Process Solution Changes

The off-gas scrubber solution (1.0 molar sodium hydroxide) was modified by adding 0.1 molar sodium thiosulfate. The thiosulfate was added to improve the ability of the solution to remove volatile iodine species. This change increased the iodine removal efficiency of the scrubber from 90 per cent to 97 per cent.

The relative effect of a rather minor change in the iodine removal scrubber solution was so great that a laboratory study of several potential scrubber solutions was made. This study is described in the Appendix. It was concluded that the most effective solutions were the sodium hydroxide-sodium thiosulfate solution in use, 1 molar silver nitrate, and carbon tetrachloride. Subsidiary process considerations made the sodium hydroxide-sodium thiosulfate solution the preferable of these three. The major objection to this solution was the formation of carbonate from contact with the air in the off-gas. The carbonate was entrained in the off-gas and sometime during each run plugged the remotely located rotameter. Studies at Hanford had indicated that 5 per cent nitric acid containing 0.001 molar mercuric nitrate and 0.001 molar mercurous nitrate was a very efficient iodine scrubbing solution (2,5). This solution, which has a very low solids content, reacts with iodine to form complex ions of mercuric or mercurous iodide (12). This solution was substituted for the caustic-thiosulfate scrubber solution. The iodine removal efficiency was not adversely affected and the rotameter plugging was eliminated.

Mercury salts were also added to the nitric acid used to neutralize the sodium hydroxide-sodium aluminate waste solution. The quantity of iodine volatilized from the solution was significantly reduced. Mercuric nitrate was also added to the acid solution of the uranium-fission product mixture. The predicted iodine retention without product contamination was observed (6,8).

IV. SUPPLEMENTARY IODINE REMOVAL FACILITY

The modifications of the original iodine removal system described in the preceding section lowered the iodine concentration in the off-gas by a factor of about 10. This reduction was sufficient to permit the safe release of RaLa off-gas during favorable atmospheric diffusion conditions, but it was not sufficient to allow RaLa operations to proceed without regard to the weather. The possibility of a sudden weather change during a RaLa run left a potential for an excessive radioactive iodine release to the environment. The addition of a supplemental iodine removal facility, in series with the original system, was the preferred solution to this problem.

Solid media for iodine adsorption were tested during the laboratory study of scrubber solution efficiencies, described in the Appendix. Two of the solid media tested, silver nitrate coated packing and activated charcoal, were superior to any of the liquids tested. It was decided, therefore, that the supplementary iodine removal facility should be a solid adsorption unit.

A. Development of Design Data

The laboratory studies of the solid adsorption media are discussed in the Appendix. Silver nitrate coated packing was eliminated because of the elevated operating temperature necessary and because of the complex regeneration procedure required. Activated charcoal appeared to be almost ideal for the proposed service. The capacity of the charcoal for iodine (as the iodine species actually present in the off-gas) was determined to be at least 80 curies of iodine-131 per cubic foot of charcoal. The admittedly sketchy regeneration tests indicated no problems.

B. Design of Plant Unit

A maximum iodine retention capacity of 500 curies of iodine-131 was assumed for sizing the adsorption unit. (The maximum iodine release actually observed was 350 curies, during a run made before all the modifications discussed in Section III were in effect.) Two adsorbers were installed in parallel, to allow regeneration of an expended adsorber while the second adsorber was on stream. Regeneration was expected to be necessary, not so much for iodine removal as for removal of entrained solids filtered from the off-gas by the charcoal beds.

Each adsorber was sized to hold 7 cubic feet of charcoal. At an iodine capacity of 80 curies per cubic foot, this size is slightly larger than required to hold 500 curies of iodine-131, but the possibility of gas channeling or solids blinding dictated the provision of a margin of safety. The overall dimensions of the adsorbers were set by the free space available for installation. Vessels 14 inches in diameter and 7 feet high, shielded with 4 inches of lead, were selected as the optimum

configuration that would fit the available space. The superficial velocity with a 14-inch diameter bed was equivalent to that used in the 1-inch diameter test beds.

Porous stainless filters (20 micron mean pore size) were installed in the outlet of the adsorbers to retain charcoal fragments or other entrained particles. Pressure differential indicators were placed across the filters and across the charcoal beds to indicate any system plugging. Victoreen radiation detection probes, connected to indicator-alarm units, were installed at the inlet and outlet of each charcoal bed to detect any radioactivity breakthroughs.

C. Performance Evaluation of Plant Unit

The iodine removal efficiency of the charcoal adsorption beds was determined by sampling the gas streams entering and leaving the beds. The samples were taken through a cartridge containing a paper filter and a section of activated charcoal. The location of the sample points and the cartridge construction are shown in Figure 1. Analyses of the cartridges showed 38 times as much iodine-131 activity retained by the inlet stream cartridge as retained by the outlet stream cartridge, indicating a 97.4 per cent removal efficiency. The cartridges were then disassembled, and the filter papers and charcoal were examined separately. Analyses of the inlet and outlet cartridge filter papers indicated an 88 per cent iodine removal, but the charcoal bed analyses indicated a 99.99 per cent removal of iodine. This apparent discrepancy may be explained on the basis that the charcoal beds were extremely efficient for the adsorption of gaseous iodine, but less efficient for the filtration of iodine-bearing particles. The overall removal of iodine from the off-gas was considered satisfactory.

The readings of bed activity obtained from the Victoreen radiation monitors on the adsorber inlets and outlets, and the analyses of stack gas iodine content during the run, generally confirmed the results obtained from the cartridge samplers.

Figure 1 - Location and Composition of Test Cartridges
Used to Determine Adsorber Performance.

V. CONCLUSIONS

Changing the chemical composition of original RaLa process solutions, modification of equipment, and revision of operating procedures made about a ten-fold reduction in the quantity of radioactive iodine released to the atmosphere. Most of this reduction probably resulted from the addition of a small quantity of mercury salts to the acidic process and waste solutions.

The charcoal adsorption units installed in series with the original iodine removal system were extremely efficient for gaseous iodine removal, but less efficient for solid particulate removal. An additional iodine release reduction of about ten-fold was obtained by using the charcoal beds.

The factor limiting the performance of a high-efficiency iodine removal unit appears to be the ability of the unit to remove iodine-bearing dusts and mists. Gaseous, or elemental, iodine can be removed with a very high (over 99.9 per cent) efficiency.

VI. ACKNOWLEDGEMENTS

The contributions of S. F. Fairbourne and P. N. Kelly to the plant and laboratory investigations and of W. B. Kerr to the design of the charcoal adsorption units were a major factor in the successful completion of this project.

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VIII. APPENDIX

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EVALUATION OF IODINE REMOVAL MEDIA

A. Liquid Media

A series of tests was made to find an improved RaLa off-gas scrubber solution. Six liquids were tested; in order of decreasing iodine removal efficiency, these were:

1. Carbon tetrachloride
2. 1M AgNO_3
3. 1M NaOH -0.1M $\text{Na}_2\text{S}_2\text{O}_3$
4. 0.1M $\text{Na}_2\text{S}_2\text{O}_3$
5. 1M NaOH
6. 2 per cent NaOCl

The iodine removal efficiency of the test solutions was determined with a side-stream of actual RaLa off-gas as the iodine source. The off-gas sample was split in parallel streams, and each stream was scrubbed in a train of gas dispersion bottles containing one of the test liquids. The flow of off-gas was adjusted to 5 scfh through each of the parallel trains. Each 500-milliliter dispersion bottle was filled with 350 milliliters of test solution; two to six bottles installed in series in each train.

It was not practical to build a test system with enough parallel test trains to include all of the test solutions in each run. To compensate for variations in the RaLa off-gas composition, a reference solution was included in each run. The sodium hydroxide-sodium thiosulfate solution was selected as the reference on the basis of its high efficiency exhibited in preliminary tests. The use of a series of dispersion bottles provided a means of regulating the extent of iodine removal and an indication of the effect of iodine content on removal efficiency.

The iodine removal efficiency of the solutions was determined by measuring the radioactivity accumulated in each dispersion bottle. Table I presents the results of a series of 5-hour test runs. The results from check runs varied slightly, as a result of variations in the RaLa off-gas, but the relative efficiencies of the solutions, listed in order previously and supported by the data in Table I, were constant from test to test.

TABLE I

IODINE ABSORPTION CHARACTERISTICS OF LIQUID MEDIA

Test	Line	Absorbent	Iodine Absorbed, Microcuries per Hour					
			Absorber Position in Line					
			1	2	3	4	5	6
1	1	1.0M NaOH-0.1M Na ₂ S ₂ O ₃	66.7	28.0	18.8	17.3	12.9	13.2
	2	1.0M NaOH	12.5	8.6				
	3	2% NaOCl	32.3	16.2				
2	1	1.0M NaOH-0.1M Na ₂ S ₂ O ₃	87.4	38.4	16.9	17.4	20.6	20.6
	2	0.1M Na ₂ S ₂ O ₃	41.9	36.6				
	3	CCl ₄	147.	0.1				
3	1	1.0M NaOH-0.1M Na ₂ S ₂ O ₃	411.	61.1	37.1	37.3	26.3	25.1
	2	1.0M AgNO ₃	596.	0.1				

B. Solid Media

During the iodine removal tests of liquid media, the effectiveness of representative solid media was also investigated. The solid media tested were (1) activated charcoal, (2) silver-nitrate-coated packing, (3) copper turnings and (4) Tygon Raschig rings. The first two of these solids were markedly superior to any of the liquid media tested that might have process application; therefore, the solid media tests were expanded to obtain the data necessary to design a plant-scale, solid-media, iodine-adsorption unit.

The relative efficiencies of charcoal and silver-nitrate-coated packing were virtually identical, preventing a choice between them on the basis of process performance. Silver nitrate reactors had the advantage of having been used for iodine removal from process off-gas at the Hanford Atomic Products Operation, with measured efficiencies of 96 to 99.5 per cent (3, 10). Silver nitrate reactors also had the serious disadvantages of requiring an elevated operating temperature (190° to 200°C) and requiring regeneration with saturated silver nitrate solutions (4). Less complete information was available on the use of activated charcoal for removal of iodine from gas streams (7, 11).

The process problems associated with the design and operation of silver nitrate reactors resulted in a decision to concentrate on the development of the simpler activated charcoal unit.

A review of the available literature indicated that an activated coconut charcoal would be preferable for iodine adsorption service. The charcoal selected for testing, and for ultimate use in the plant adsorbers, was a 1.88 millimeter diameter extruded charcoal, Norit R-2, with a 4 to 14 mesh Tyler screen analysis. The charcoal was supplied by L. A. Salomon and Bro., New York.

The test setup used to determine the iodine removal characteristics of activated charcoal was very similar to that used for the liquid media tests. A side-stream of RaLa process off-gas was passed through charcoal beds and then through a carbon tetrachloride scrubber. The carbon tetrachloride was assumed, on the basis of the tests reported in Part A, to collect any iodine not removed by the charcoal. Tests were made with the charcoal beds in series and in parallel, depending on the data required from each particular test. Two charcoal cartridge sizes were used; one was 0.038 inches inside diameter, containing 2 grams of charcoal, the second was 1.049 inches inside diameter (1-inch, Schedule 40 pipe), with a varying depth of bed. The smaller cartridges were only used for preliminary tests, because of the danger of wall effects that could obscure the test results.

The primary test involved the use of four 1-inch diameter charcoal beds in parallel. Bed depths for the four cartridges were one inch, two inches, four inches, and eleven inches. The flow through each bed was 5 scfh. Samples were taken from the carbon tetrachloride scrubber following the one-inch-deep bed every 30 minutes to detect the iodine breakthrough point. The test was continued for the full 24-hour duration of the RaLa run, but no breakthrough was reached. At the end of the run, the iodine contents of the charcoal beds and carbon tetrachloride scrubbers were measured by the iodine-131 activity levels.

The iodine removal efficiency of the charcoal beds increased with increasing bed depth: 93 per cent for a 1-inch bed, 96 per cent for a 2-inch bed, 96.5 per cent for a 4-inch bed, and 97.4 per cent for an 11-inch bed.

Since no iodine breakthrough was detected during the test run, the 1-inch-deep bed was dissected to provide an estimate of the approach to breakthrough. The bed was removed as 33 successive layers, and each layer was analyzed separately. The analytical results are shown in Figure 2. Although the data show some scatter, it is obvious that the iodine-retention capacity of the charcoal bed was not approached during the test. The average iodine content of the 1-inch bed was 80 curies of iodine-131 per cubic foot of charcoal. This value was used as the iodine capacity of the charcoal to design the plant adsorber units, but the data in Figure 2 indicate this value is conservative by at least a factor of two.

The failure of the longer carbon beds to remove essentially all of the iodine and the deviation of Figure 2 from typical adsorber performance may both be explained on the basis of one postulate. A significant portion of the iodine appears to be present in the RaLa off-gas in the form of particles. Verifying evidence was obtained from further analyses. The radioactivity retained on the last of four charcoal beds in series was about 60 per cent from iodine-131 and about 40 per cent from radioactive isotopes of barium, lanthanum, zirconium, niobium, and ruthenium. These latter elements, with the possible exception of ruthenium, must have been carried in the off-gas as a dust or mist.

The presence of entrained particulates in the off-gas was also confirmed by the spectroscopic detection of sodium on the charcoal at the bed inlet position. It is reasonable to suppose that this entrainment would contain iodine compounds, and that these compounds would be removed only by the filtration, rather than adsorption, properties of the charcoal beds. This supposition offers a very lucid explanation of the observed tendency for iodine to "smear" through the charcoal beds. It also indicates that the upper limit of iodine efficiency will be determined in practice by the filtration efficiency of the unit employed.

Regeneration of expended charcoal was not expected to be a problem, and relatively little work was done on regeneration procedures. Charcoal bed regeneration by steam or air is a routine operation. In this particular case, had any severe problems arisen in the removal of collected iodine, the short half-life of iodine-131 meant that in a fairly short time the iodine would be removed by radioactively decaying to the noble gas xenon.

Used charcoal test beds were regenerated with steam and with heated air, and reused in parallel with beds of fresh charcoal. So far as could be determined, the performance of the regenerated charcoal was identical to that of fresh charcoal. No attempt was made to determine the ultimate capacity of the charcoal; the time required to complete the necessary number of RaLa runs would have been prohibitive.

Figure 2. Incremental Activity of Charcoal Layers from Test Bed Exposed to RaLa Process Off-Gas.

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