Control of Radio-Iodine at the German Reprocessing Plant WAK during Operation and after Shutdown

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Abstract

During 20 years of operation 207 metric tons of oxide fuel from nuclear power reactors with 19 kg of iodine-129 had been reprocessed in the WAK plant near Karlsruhe. In January 1991 the WAK Plant was shut down. During operation iodine releases of the plant as well as the iodine distribution over the liquid and gaseous process streams had been determined. Most of the iodine is evolved into the dissolver off-gas in volatile form. The remainder is dispersed over many aqueous, organic and especially gaseous process and waste streams.

After shut down of the plant in January 1991, iodine measurements in the off-gas streams have been continued up to now. Whereas the iodine-129 concentration in the dissolver off-gas dropped during six months after shutdown by three orders of magnitude, the iodine concentrations in the vessel ventilation system of the PUREX process and the cell vent system decreased only by a factor of 10 during the same period. Iodine-129 releases of the liquid high active waste storage tanks did not decrease distinctly.

The removal efficiencies of the silver impregnated iodine filters in the different off-gas streams of the WAK plant depend on the iodine concentration in the off-gas. The reason of the observed dependence of the DF on the iodine-129 concentration might be due to the presence of organic iodine compounds which are difficult to remove.

I. Introduction

Besides the optimization of the PUREX process particularly by an electrolytic uranium-plutonium separation, an important task for the WAK plant was to demonstrate low iodine releases in view of the project of the German industrial scale reprocessing plant.

In 1975 the yearly iodine-129 release limit of the WAK plant via the off-gas was fixed to 240 MBq (equivalent to 37 g iodine-129). By this reason silver impregnated iodine filters had been installed in the dissolver off-gas.

Main objective of the iodine management during operation of the WAK plant was to obtain a low residual iodine content in the fuel solution after the end of the dissolution process by evolving most of the iodine into the dissolver off-gas and an almost quantitative iodine removal on silver impregnated solid bed filters in the dissolver off-gas. At the beginning of 1988 silver impregnated solid bed iodine filters entered in operation in two vessel ventilation systems.

In the meantime, the licensing authorities had reduced the maximum permissible iodine release of the planned German industrial scale reprocessing plant in Bavaria from 7.4 GBq (200 mCi) to 1.85 GBq per year (50 mCi) for a throughput of 350 tons of uranium from light water reactor fuel (burnup 50,000 MWd/ton U). This value of 1.85 GBq signified that only 0.2 % of the iodine inventory could be released by all off-gas streams of the plant.

After some years of intensive work at the Karlsruhe reprocessing plant WAK in 1989 it was clear that this ambitious aim of 0.2 % release of the iodine inventory could not be demonstrated in the WAK plant. Because of the shortness of time before shut down there was no possibility to install iodine filters in all off-gas lines of the WAK plant. Meanwhile the project of the industrial scale reprocessing plant in Germany had been given up in 1989 after 15 years of intensive development work.

II. Iodine Distribution in the WAK Plant

During fuel dissolution the fission product iodine is present mainly in its volatile elementary form. From the point of view of radio ecology, iodine-129, with a half life period of $1.6*10^7$ years and iodine-131 with a half life period of 8.02 days, are significant. If cooling times are > 1 year, as in the case of commercial reprocessing, practically all of the short life I-131 from uranium fission has decayed.

However, iodine-131 from fission of curium-244 in the fuel is detected and measured in all steps of the PUREX process: in the dissolver off-gas, in the vessel ventilation system, and particularly in the vessel ventilation of the storage tanks of the liquid high active waste (Herrmann 1994).

II.1 Residual Iodine Content in the Dissolver Solution

The chemical behavior of iodine in a nitric acid medium is complex. Particularly at low concentrations, iodine behavior is dominated by the redox potential in combination with unavoidable, mostly organic trace impurities in the solution. The species and the concentration of these impurities depend on the suppliers of nitric acid and on processing details. So the behavior of iodine may deviate largely from expected textbook behavior (Henrich 1996, Comor 1992).

The conditions for evolution of iodine into the off-gas have been examined in laboratory, in inactive pilot plant scale, in the hot cell facilities of the Karlsruhe nuclear research center as well as in the WAK reprocessing plant.

Henrich, Grimm and Boukis (1991) demonstrated with a high burnup fuel (50 GWd/T U) from the BIBLIS light water power reactor in laboratory scale dissolution experiments residual iodine contents between 1.1 % (1.2 mg/l) and 2.2 % (2.3 mg/l) of the inventory of the fuel after dissolution of 80 g of UO_2 fuel in 220 ml HNO_3 (7.5 M). The values of 1.2 mg/l and 2.3 mg/l have been obtained with nitric acid of p.a. quality respectively with recycled acid from the WAK plant (Henrich 1991). After filtration of the fuel solution, addition of iodate carrier, subsequent NOx sparging and a second filtration of the fuel solution the residual iodine content decreased below 0.5 % (0.5 mg/l) of the iodine inventory of the fuel. The residual iodine concentration depends strongly on the quality of the nitric acid.

Because of the extraction behavior of the residual iodine in different extractants, Henrich (1991) and Boukis (1991) conclude that the residual iodine must be a mixture of many iodinated organic species with different extractabilities. Henrich (1988) pointed out that the concentration of organic impurities in a pure nitric acid in a ppm range is by two orders of magnitude higher than the aspired residual iodine content in the dissolver solution ($2*10^{-6}$ mol/l). Henrich's interpretation is supported by the results of Lieser (1989).

The WAK plant experiences (>8 hours boiling time, air sparging, addition of carrier iodine during the last 2 hours of dissolution) showed, that it is difficult to reach residual iodine values in the dissolver solution below 1 % of the inventory for a high burnup fuel (35,000 MWd/T U). This value corresponds to an iodine concentration of approximately 1 mg/l.

For dissolution in WAK lower iodine-129 concentrations than 1 % of the iodine inventory in the feed have been reported by Berg (1977). The discrepancy between the earlier WAK values and the recent values of the Karlsruhe research center and of the WAK plant is not connected with the evolving process but with the usual analytic iodine separation and determination procedure which took into account only the inorganic and soluble iodine compounds (Henrich 1996).

Depending on operation conditions in the WAK plant (8 hours boiling time, air sparging), the residual iodine concentrations in the dissolver solution generally were in the range of 1 to 4 mg iodine per liter. It seems that the residual iodine concentration [g/l] is not influenced by the burnup of the processed fuel; the fraction of the residual iodine in [%] is of course higher for the fuel with a low burn-up and with a low total iodine content. Fuel with a low burn-up of 7 to 14 GWd/t U yielded residual iodine contents of 4 % to 12 % (in average 6 %), whereas high burn-up fuel with 30 GWd/t U yielded average residual iodine contents of 2 to 3 % of the inventory of the fuel after the end of the dissolution. In both cases, high burn-up fuel and low burn up fuel, the range of the residual iodine concentration was 1 to 4 mg/l. (Fission product iodine-127 was taken into account with 25 weight % of the iodine-129 inventory).

Important parameters for the evolution of the iodine during the dissolution process are the quantity of water vapor generated during boiling of the dissolver solution (45 m^3/h), the formation of the nitrous oxides (20 m^3/h) and the duration of the dissolution process. For the standard dissolution conditions 8 hours are required. To obtain low residual iodine values in the dissolver solution it is important to have a high quantity of dissolver off-gas passing through the dissolver and the reflux condenser (80 to 120 m^3/h). The addition of natural iodine as carrier and sparging of the dissolver solution with NO_2 during the last hour of the dissolution resulted in an average iodine content of 1 % (about 1 mg/l) of the iodine-129 inventory in the case of a high burnup fuel.

The percentages employed for the following considerations are average values resulting from the analyses of many liquid and many off-gas samples. These values cover the analyzed iodine fractions obtained by fuel with burn-up between 15,000 MWd/t U and 40,000 MWd/t U.

II.2 Iodine Concentration in the Dissolver Off-Gas

In the Karlsruhe reprocessing plant the bulk of the iodine inventory (in average 94 %) is evolved from the dissolver solution and is transported by the dissolver off-gas DOG through a reflux condenser and two subsequent acid scrubbers on two iodine filters in series. These filters contain a silver nitrate impregnated silica material with a grain size of about 1 mm. The iodine concentration in the dissolver off-gas upstream the iodine filters undergoes big variations. Average values during dissolution periods are in the order of 1 to 5 mg/m^3 with peak values up to 100 mg/m^3.

After shut down of the WAK plant the iodine concentrations upstream the iodine filter in the dissolver off-gas dropped from average values of about 2 mg/m^3 in December 1990 to 0.03 mg/m^3 in January-February 1991 and further to 0.001 mg/m^3 at the end of 1991.

II.3 Iodine Depositions in the Sampling Lines of the Dissolver Off-Gas

Iodine depositions in sampling lines have been studied by Glissmeyer and Sehmel (1991). They occur in the sampling lines because of the high surface/volume ratio of the sampling pipes and the relatively low linear velocity of the sampling gas streams. Iodine depositions in the sampling lines can potentially lead to erroneous results of radioiodine measurements and make dubious all short term measurements concerning the removal efficiency of iodine filters or scrubbers. One example may illustrate the problem of iodine depositions:

Iodine sampling in the dissolver off-gas of WAK is performed discontinuously all 4 to 5 dissolutions. At the beginning of a dissolution campaign in WAK after 5 dissolutions of a fuel with a burnup of 14,000 MWd/t U only about 34 % of the iodine inventory of the fuel could be measured in the dissolver off-gas upstream the iodine filters. (This value can lead to the conclusion that the scrubbers upstream the iodine filters had an iodine removal efficiency of 60 %, assuming an iodine evolution of 94 % of the iodine inventory in the dissolver). After 14 dissolutions about 76 %, and after 32 dissolutions about 84 % of the iodine inventory were found on the sampling filters. These delayed iodine concentrations result from iodine depositions formed in the sampling lines at the beginning of a campaign.

The above cited results had been obtained without heating the sampling lines. To overcome the errors due to iodine depositions in non heated sampling lines, long term measurements have to be performed. By heating the whole sampling lines to > 80 °C, it had been demonstrated that iodine depositions can be prevented and that short term measurements of the removal efficiency of the iodine filters yield reliable results.

After the end of a dissolution period iodine concentrations in the dissolver off-gas system drop slowly. Several days are necessary to attain iodine concentrations comparable to the iodine concentrations in the vessel ventilation system of the PUREX process (0.008 mg/m^3).

II.4 Iodine Behavior in the PUREX Solvent Extraction Process

The percentages of iodine in the following considerations are average values resulting from the last five years of operation. They cover the analyzed iodine fractions obtained by fuel with burn-up between 15,000 MWd/t U and 40,000 MWd/t U (see chapter II.1).

The residual iodine species remaining in the fuel solution (in average about 5 % of the iodine inventory of the fuel; about 1 % is fixed on the dissolver residues) are distributed in the subsequent extraction process operations. About 2 % to 3 % of the iodine inventory of the dissolved fuel were found in the organic solvent (20,000 to 40,000 Bq iodine-129/l corresponding to 3 mg/l to 6 mg/l). The rest of about 2 % are spread over the high active waste and different off-gases. The organic iodine compounds in the solvent contribute to the iodine emitted by the vessel vent system of the PUREX process and the vessel vent of the subsequent medium active waste treatment facility. During solvent wash the iodine content in the organic phase does not decrease considerably. Only about 0.3 % of the iodine inventory of the fuel was found in the alkaline solvent wash streams.

After shut down of the WAK plant the organic solvent, the organic and aqueous wastes have been transferred to the waste treatment facilities of the Karlsruhe Research Center. The iodine releases of this facility had been discussed by Amend (1990).

II.5 Iodine Concentration in the Vessel Ventilation System of the PUREX Process

After 8 hours of dissolution the residual iodine concentration in the dissolver solution is 1 to 4 mg/l. The residual iodine is routed to the extraction process. In 1990 detailed investigations over different reprocessing periods were carried out during reprocessing of about 9 tons of fuel from nuclear power plants (burnup 5.4 to 30 GWd/t) to determine the distribution of iodine in the PUREX process. From the iodine arriving in the separations process, about 50 % are evolved into the vessel vent system. The average iodine concentration in the vessel vent (270 - 330 m^3/h) is between 0.008 and 0.018 mg/m^3. Maximum values measured over a 14 days period attained about 0.03 mg/m^3 (160 Bq/m^3 Iodine-129).

Immediately after shut down the iodine release via this off-gas dropped from about 10 MBq (1.5 g of iodine-129) per month in December 1990 to 5 MBq per month in January 1991 and reached values of about 0.2 MBq/month (= 30 mg/month) after 10 months.

II.6 Iodine in the Vessel Ventilation System of the Medium Active Waste Storage Tanks

The iodine concentrations in this off-gas stream during operation was approximately 0.01 mg/m^3. After shut down this concentration did not drop immediately. One year after shut down the iodine concentration was lower by a factor of 10. No iodine filter was in operation in this off-gas.

II.7 Iodine in the Vessel Ventilation System of the Liquid High Active Waste Concentrate (HAWC) Storage Tanks

Two storage tanks contain the high active waste concentrate of 20 years reprocessing history of the WAK plant (about 60 m^3 HAWC).

During the operation period of WAK the iodine concentration measured in this off-gas stream was between 0.002 to 0.006 mg/m^3. (iodine-129: 10 to 30 Bq/m^3).

After shut down of the plant in January 1991 the monthly amount of iodine-129 evolved by the HAWC has not decreased considerably during five years. In average 2 MBq of iodine-129 per month (0.3 g I-129) are evolved. Table 1 gives an overview over the iodine-129 evolved.

Year	1991	1992	1993	1994	1995
I-129 evolved [MBq/a]	16.9	16.1	23.7	25.8	26.5

Table 1 Iodine-129 evolution from the liquid high active waste concentrate (HAWC)

The amount of iodine-131 evolved by the HAWC solution in terms of Becquerel is in the same order of magnitude as the amount of Becquerel of iodine 129. Iodine-131 results mainly from the spontaneous fission of Curium-244. Whereas Cm-244 and Cm-246 in the waste solution are the main sources for the generation of iodine-131 by spontaneous fission (Herrmann 1994), the iodine-129 evolved by this off-gas results mainly from the residues of the dissolution processes from 20 years reprocessing, containing some hundred grams of iodine-129.

Up to now the evolution of iodine-129 from these storage tanks show no significant decrease. Due to the iodine filter in this vessel vent system the release values are about 10 % of the amount of the iodine evolved by these solutions.

II.8 Iodine in the Cell Ventilation System

In the cell vent system of the WAK plant during dissolution periods a fraction of 0.3 % of the iodine-129 inventory of the fuel was measured. Iodine monitoring devices in different ventilation systems revealed that the measured iodine (up to 30 mg per day) originated almost exclusively to the shear and dissolver cell. The measured iodine concentrations in this off-gas were $3*10^{-5}$ mg/m³ and reached concentrations up to $18*10^{-5}$ mg/m³ during plant operation. Such high release values have been observed exclusively during dissolution periods. The high releases of 20 to 30 mg of iodine per day had been measured by two independant monitoring systems over a period of 4 to 6 weeks. It can be excluded that the measured iodine was a result of the shearing process itself. Leudet and Leseur (1982) demonstrated that during shearing of high burnup fuel from power reactors fission product iodine release was not detectable.

Two reasons may be responsible for an iodine leaking out of the dissolver:

- A leakage of the dissolver cover seems to be possible at the beginning of dissolution when a high rate of nitric oxides are generated and the reduced pressure of the dissolver off-gas system goes to 0 (related to the pressure in the dissolver cell).
- The reduced pressure cannot be maintained during the opening of the dissolver cover when the fuel is loaded and iodine deposits may come into the cell.

After shut down the iodine-129 release values dropped from about one MBq/month in December 1990 to 0.1 MBq/month in November 1991.

III. Iodine Removal Devices

III.1 Acid Scrubbers and Caustic Scrubber in the Dissolver Off-Gas

Because of the presence of nitric oxides in the off-gases of a reprocessing plant the use of charcoal is prohibited.

Acid scrubbers: Concerning the two acid scrubbers in series located in the dissolver off-gas system of the WAK plant only 0.5 % to 1.5 % of the iodine inventory of the dissolved fuel had been measured in the nitric acid solution of the two packed columns after each dissolution charge. This fact of a low removal efficiency must be seen in connection with the relatively high dissolver off-gas flow rate of the WAK plant of about 100 m³/h.

Caustic scrubber: At the beginning of 1975 during WAK operation it has been tried to remove the iodine evolved from the dissolver by caustic scrubbing. An iodine removal efficiency of 53 % to 71 % could be attained in the dissolver off-gas (Schüttelkopf 1976). Because this iodine removal efficiency was too low, iodine filters with silver impregnated silica entered in operation mid 1975 in the dissolver off-gas. In the vessel ventilation system only 15 % of the total measured iodine could be removed by caustic scrubbing (Herrmann 1990).

III.2 Silver impregnated Iodine Filters in the Dissolver Off-Gas

During dissolution periods the highest removal efficiencies for the two filters in series of 99.5 to 99.9 % related to the iodine concentrations in the off-gas upstream the iodine filter had been measured, the iodine concentrations in the dissolver off-gas varying in average between 1 and 5 mg/m^3. Downstream the two iodine filters in series iodine-129 concentrations of about 0.005 mg/m^3 were observed during dissolution periods.

During outage periods and during the first six months after shut down, iodine concentrations of about 0.01 mg/m^3 (65 Bq/m^3) had been measured upstream the two iodine filters. Downstream the iodine filters iodine concentrations of about 0.0007 mg/m^3 were measured. The removal efficiency of the two iodine filters in series dropped to 90 %.

One year after shut down the iodine-129 concentrations downstream the two iodine filters dropped to about 0.6 Bq/m^3.

III.3 Silver Impregnated Iodine Filter in the Vessel Off-Gas of the PUREX Process

As shown above the iodine concentration in the vessel off-gas varied between 0.008 and 0.018 mg/m^3. The measured removal efficiency of a prototype filter in a side stream of this off-gas has been 90 to 95 % of the iodine concentration in the off-gas upstream the iodine filter.

III.4 Iodine Filter in the Vessel Ventilation System of the High Active Waste Storage

This iodine filter with AC6120/12 % Ag is operating at about 40 °C and has a removal efficiency 90 % to 94 %. The gas flow rate is about 180 m^3/h. The measured iodine contents in the off-gas upstream (evolved) and downstream (released) are shown in table 2.

Year	1990	1991	1992	1993	1994	1995
Iodine-129 evolved [MBq/a]	35.0	16.9	16.0	23.7	23.9	26.5
Iodine-129 released [MBq/a]	2.3	0.9	1.5	2.0	1.72	1.4

Table 2 Iodine contents upstream and downstream the iodine filter in the vessel ventilation system of the storage tanks of the liquid high active waste concentrate (HAWC)

IV. Iodine Release of the Reprocessing Plant WAK during Operation and after Shut Down

The iodine release fraction of a reprocessing plant is defined as the ratio of all iodine leaving the plant to the iodine processed in the plant. It is determined by the efficiency of the iodine removal devices.

Most of the iodine released resulted from the vessel ventilation systems. During the considered period the vessel ventilation system had no iodine filters. Although the iodine removal efficiency of the filter in the dissolver off-gas was high (about 99.5 %), the iodine overall release from the plant was in yearly average 1.8 % of the iodine inventory of the fuel during 14 years of reprocessing. The highest release fraction in one year was 4.1 % of the iodine processed. This high release fraction resulted from an experimental program with the aim of shortening the dissolution time of 7 dissolution charges from 8 hours to 5, 4 and 3 hours. It must be noted that this experimental program concerned only 7 from 25 dissolution charges in one month.

The values of iodine 129 releases during the last year of operation (1990) and the following years after shut down demonstrate clearly a decrease of the iodine-129 concentration by a factor of about 10 in the off-gases.

Table 3 shows the iodine-129 and iodine 131 releases of the WAK Plant from 1990 to 1995.

Year	1990 Operation	1991 shut down	1992	1993	1994	1995
Iodine-129 (MBq/a)	91	33	8.7	6.5	4.3	2.9
Iodine-131 (MBq/a)	12	3.7	4.5	4.5	4.1	2.5

Table 3 Iodine release of WAK Plant from 1990 through 1994 (shut-down January 1991)

After shut down most of the iodine releases is originated from the vessel vent system of the medium active waste storage tanks and the high active waste concentrate (HAWC) storage tanks. The HAWC evolves in average 25 MBq iodine-129 per year. Since this off-gas stream passes through an silver impregnated iodine filter, the releases are lowered by a factor of about 10.

The iodine 131 emissions after shut down result exclusively from the high active waste concentrate. The amount of iodine-131 evolved shows a slight tendency of decrease during the last five years due to the half life period of the nuclide Cm-244 (18 years), generating the iodine-131.

During the last year of reprocessing (1990) the release fraction of the WAK plant for iodine-129 was 2.1 % of the iodine-129 inventory processed. From this 2.1 % two thirds originated to the vessel ventilation system of the PUREX process, one fifth to the storage tanks of the medium active waste and one tenth to the cell ventilation system. Only a fraction of 0.028 of the 2.1 % originates to the iodine filtered dissolver off-gas and a fraction of 0.018 to the iodine filtered vessel ventilation system of the liquid high active waste concentrate storage tanks.

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DISCUSSION

SAKURAI: At the WAK reprocessing plant, was the expulsion of iodine from the spent-fuel solution performed by NO_x sparging? And what was the effect?

HERRMANN: It was an experimental program which took a lot of preparation because we must make the demand to the authorities. But we have already made ten dissolutions with additions of I-127 and NO_2. NO_2-sparging and simultaneous addition of iodine-127 to the dissolver resulted in a residual iodine content in the feed solution of 1 mg/l.

JUBIN: I have a question on the high active waste concentrate. Have you had a chance to analyze the iodine content in the HAWC?

HERRMANN: The problem with the iodine-129 analysis was the biggest problem we had because the analytical procedure, by extracting and reextracting the iodine from the high activity solutions did not catch the organic species. And as far as I remember, in the high active waste solutions, we found between 500 and 5,000 B8/l before the concentration step (HAWC). But this was not compatible with the continuous evolution of iodine-129 tanks. (The measurement of iodine-129 in the offgases is reliable because after the HEPA filters, there is no other activity on these sampling filters.) The continuous evolution of I-129 from HAWC results from the solid residues in the HAWC containing iodine.

KNOCH: Iodine depositions can occur also in the main ducts of the offgas.

HERRMANN: In our case the linear velocity in the sampling lines is lower than the gas velocity in the main line. I agree, we cannot exclude depositions in the main pipes.

FUKASAWA: In Table 3 of your manuscript, I -129 release decreased more rapidly than I-131. (I-129: 91 MBq/y ('90) ⇨2.9 ('95); I-131: 12 ('90)⇨2.5 ('95) What is the reason for this phenomenon?

HERRMANN: The rapid decrease of I-129 releases is conspicuous after shutdown of the plant in January, 1991. The I-131 releases after shutdown result from the spontaneous fission of Cm-244 present in the high level liquid waste HAWC.