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Experiment on the Gaseous Iodine Release from Irradiated Cesium Iodide Solutions (Contract Research)

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The volatile iodine production due to radiation chemical effects in the containment vessel of light water reactors (LWRs) during severe accidents, known as an important uncertainty factor in the source term evaluation, was investigated by experiments in small scale and with well controlled conditions. Cesium iodide solutions, 10^{-4} M, labeled with ^{131}I, at controlled pH by boric acid-sodium hydroxide buffer, were gamma-irradiated and swept with a constant gas flow rate. The gaseous iodine released from the solution was collected by species selective filters and quantified separately for I_2 and organic iodines. The influences of pH, temperature, inorganic and organic impurities, oxygen and hydrogen concentrations in the cover gas on the iodine release behavior were examined. The results showed the following trends: lower pHs cause higher release fractions and the pH impact is the most significant; higher temperature or hydrogen in the cover gas cause lower release fractions; lower oxygen concentrations cause higher release fractions; addition of an organic compound (methylisobutylketone, MIBK) causes decrease of I_2 release and increase of organic iodine release, and the effect is more significant with lower oxygen concentrations; influence of iron and chloride ions were not significant.

The data on the time dependent gaseous iodine release fractions, comparison of the final iodine release fractions in terms of the parameter effects, as well as the initial, boundary and interface conditions necessary for simulating the experiments by computer codes are provided.

Two additional series of experiments on suppression of iodine release by ammonia alkaline under irradiation, and comparison of the influence of two organic compounds, MIBK and xylene, on the organic iodine production were performed. With the ammonia alkaline conditions (pH$\sim$11), iodine release fraction was small in both the irradiated and not irradiated cases, and significant difference was not detected. The organic iodine production with MIBK and xylene were similar.

Keywords: Severe Accident, Containment Vessel, Source Term, Volatile Iodine, Radiation Chemistry, Organic Compound, Oxygen, Hydrogen

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照射によるヨウ化セシウム水溶液からのガス状ヨウ素放出に関する実験 (受託研究)

日本原子力研究開発機構 安全研究センター
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(2011 年 4 月 21 日 受理)

軽水炉シビアアクシデント時の格納容器内における放射線化学的作用による揮発性ヨウ素の生成は, ソースターム評価における重要な不確かさ要因である. この現象に関する実験を小規模な体系で良く制御された条件下において実施した. ^{131}I で標識し, ホウ酸-水酸化ナトリウム緩衝剤によりpH を制御した 10^{-4} M ヨウ化セシウム水溶液にガンマ線を照射し, 一定流量のガスで掃気することにより, 気相へ放出される揮発性ヨウ素を化学種選択性のフィルタを用いて I_2 と有機ヨウ素に分別捕集し, 各々の放出量を測定した. また, ヨウ素放出挙動に対する pH, 温度, 無機及び有機不純物, 雰囲気中の酸素及び水素濃度の影響を調べた. 試験結果は以下の傾向を示した. 低 pH でヨウ素放出が増加しその影響は最も顕著である. 高温, 雰囲気中の水素はヨウ素放出の減少をもたらす. 低酸素濃度でヨウ素放出は増加する. 有機物 (メチルイソブチルケトン,MIBK) を添加すると, I_2 としての放出は減少し有機ヨウ素としての放出が増加し, 低酸素濃度では影響がより顕著である. 鉄イオン及び塩化物イオンはほとんど影響が見られない.

本報告書には, ヨウ素放出割合の時系列変化に関するデータ, 及び最終放出割合に対する各パラメータの影響に関する比較を示し, 併せて, 計算コードにより本実験のシミュレーションを行う場合に必要な初期・境界条件及び界面条件を示す.

補足実験として, 照射下でのアンモニア塩基性によるヨウ素放出抑制に関する実験, 及び, 有機ヨウ素放出に対する MIBK とキシレンの影響の比較実験を行った. アンモニア塩基性下 (pH∼11) ではヨウ素放出割合は小さく, 照射による有意な影響は見られなかった. MIBK とキシレンによる有機ヨウ素生成量は同程度だった.

本件は (独) 原子力安全基盤機構からの受託研究 (平成 19 年度 ∼ 平成 22 年度) の成果を含む.
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1 Introduction

Iodine is one of the fission products (FPs) produced in the core of light water reactors (LWRs) and is the one recognized as the most relevant to the public health consequences in case of a severe accident with environmental release of FPs, due to its large fission yield, the half life of 8 days (for ^{131}I) and complicated chemical behavior that may produce volatile species. Thus, large number of research works have been done on the iodine chemistry under the conditions relevant to the reactor cooling system (RCS) and the containment vessel (CV) of LWRs. Especially, after the core degradation accident at TMI-2 plant, US, in 1979, comprehensive studies focused on more realistic evaluation of the iodine source term have been performed internationally.[1, 2, 3]

In the RCS, iodine speciation is primarily determined by thermodynamics and CsI vapor or aerosol is the dominant form of iodine. Typically, Beahm et al. (1991)[4] concluded that the iodine at the release into the CV is mostly in CsI and less than 5% elemental iodine and HI.

In the CV where temperature is lower, iodine as CsI becomes aerosols and mostly sediments on to the floor gravitationally or by the spray, then dissolves into water as iodide ion. It is known that iodide ion is converted into volatile elemental iodine or organic iodine, under the effect of radiation. Due to the low temperature in the CV, the speciation does not follow the thermodynamics but is dominated by kinetics. Various influences on the radiation chemical reaction of iodine were investigated, such as pH, initial iodide concentration, dose rate, temperature, dissolved gases and impurities.[5, 6, 7, 8, 9] It is known that the solution pH has a strong effect, *e.g.* the yield or the release rate of gaseous I_2 increases by more than an order when the pH decreases from 8 to 5.[7, 9]

Organic impurities in the solution cause production of organic iodines. A possible major source of such organic impurities is the organic paint on the containment wall.[2, 3] Significant amount of solvents remaining in the paint matrix leach into the pool water when submerged under water, and undergo radiolytic decomposition. The resulting organic radicals react with iodine and produce organic iodines, some of which are more volatile than elemental iodine and less adsorptive on walls, thus more persistently reside in the gas phase.

We performed a series of experiments on the volatile iodine release from irradiated CsI solution with a small scale well controlled experimental apparatus. The dependence on pH, temperature, influences of inorganic and organic impurities and cover gas conditions were examined. Especially, we looked into the conditions relevant to BWRs where the containment is purged of oxygen with nitrogen for the purpose of hydrogen combustion control, since most of the recent research work in this field was more focused on PWR and CANDU plants where the atmosphere in the containment is normal air.

This report describes the methods, conditions and results of the experiments, observations on the parametric influences of the factors examined. Also, we provided the boundary, interface and initial conditions of the experimental system, to facilitate the reference of the present data for computer simulations and model verifications.

Chapter 2 describes the gaseous iodine release experiment with ^{131}I-labeled CsI solution, in which the iodine release fraction due to the aqueous radiation chemical processes during the irradiation was well quantified and parametric effects were examined comprehensively. Chapter 3 describes two kinds of experiments without ^{131}I tracer. In one series, behavior of ammonia alkaline CsI solution under irradiation was examined. In the other, influences of two organic compounds, xylene and methylethylketone (MIBK), on the organic iodine production were compared. In the Appendixes, a complementary experiment giving the interface mass transfer coefficients in the present system is described, and a collection of data for each case are provided.

2 Gaseous Iodine Release Experiment

2.1 Experimental Facility

2.1.1 System

Figure 2.1 shows the experimental facility. It is a small scale once through flow system. A glass vessel containing test solution, typically 100 mL of ^{131}I-labeled 10^{-4} M (mol/L is denoted by M) CsI solution buffered at prescribed pH and with prescribed additives, was set in a ^{60}Co γ-irradiator. The cover gas over the irradiated solution was swept with prescribed gas, and the gaseous iodine species released from the solution was transferred to the samplers set in the online counting apparatus in the downstream. The iodine release fraction was continuously measured by the ^{131}I activity accumulating in the samplers.

The release rate depends on the gas-liquid interface mass transfer and wall adsorption as well as the aqueous chemical reaction. To isolate the effect of the aqueous chemical reaction from others, we controlled the interface mass transfer by giving a constant stir and we evaluated the transfer coefficient separately by a complementary experiment (see Appendix A). We tried minimizing the adsorption of iodine in the experimental system by carefully choosing the materials of the vessel and the tubes.

The sweep gas was provided from a cylinder through a pressure regulator, charcoal and glass fiber filters (CP-20 and GB-100R, 47 mm aperture, Advantec), an over flow chamber and a humidifier at room temperature. In the downstream, the gas went through the iodine samplers, a condensate separator, silica-gel and charcoal filters, a mass flow controller, and then exhausted through a pump.

The tube connecting the irradiation vessel and the iodine sampling system was 1/4” outer diameter, made of electro-polished 316 stainless steel for about 300 mm from the vessel exit and of polytetrafluoroethylene (PTFE) for the rest. The tube was thermostated at 40–45 °C and vapor condensation was avoided.

When hydrogen containing gas was used, hydrogen detectors (Gasman H2, Crowcon) were placed near gas exits, and the hydrogen concentration relative to the lower explosion limit (LEL), 4% H_2 in air, was checked.

Description of primary components of the system is given in the following.

2.1.2 γ-Irradiator

Figure 2.2 shows the outlook of the γ-irradiator (GC220, MDS Nordion). The irradiation source is a set of ^{60}Co pins surrounding the irradiation space at the center of the main lead shield. A vertical drawer carrying an irradiation chamber at its center moves up and down, and the material set in the chamber is irradiated when the drawer is inserted in the irradiation space.

The source activity was 407 TBq as of Feb. 20, 2007. The absorbed dose rate in the test solution was determined separately by ceric/cerous chemical dosimetry[10] in the same geometric condition as in the present experiment. The dose rate varied between cases due to the decay of ^{60}Co source and ranged 5–8 Gy/h during the period of this program. The irradiation chamber has an access hole to the above. Gas tubes, electric cables and other necessary equipments were provided through it.

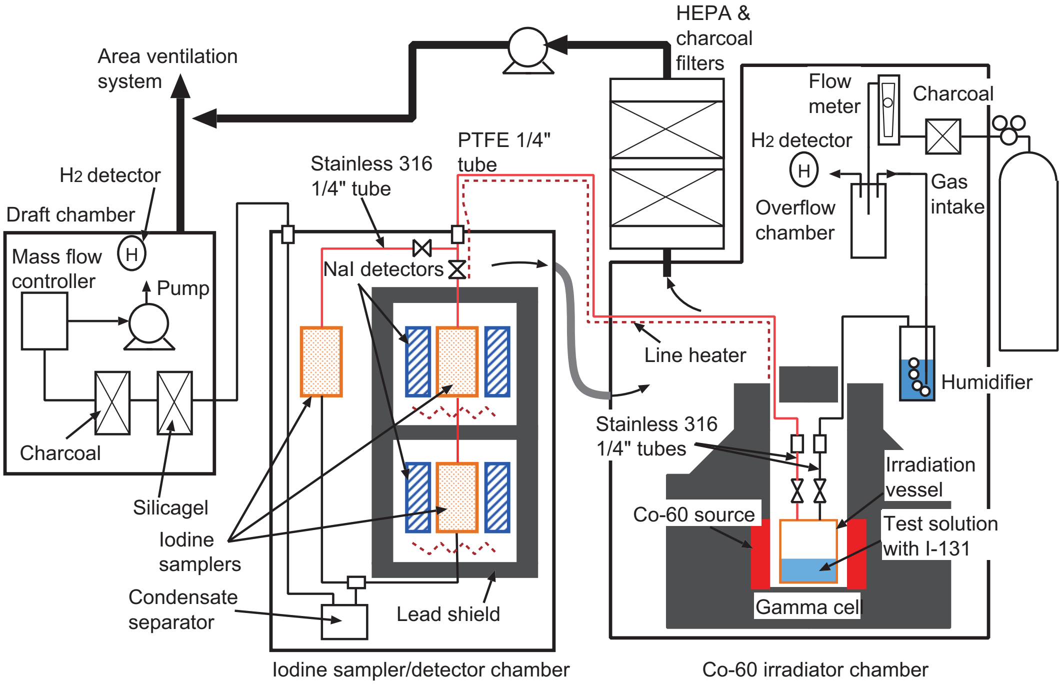

Figure 2.1: Schematic illustration of the experimental system.

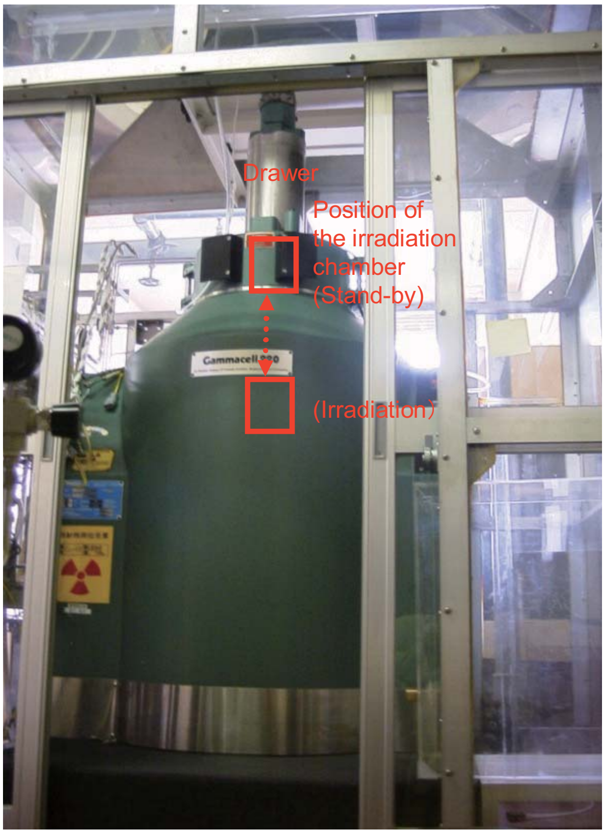

Figure 2.2: Gamma irradiator GC220.

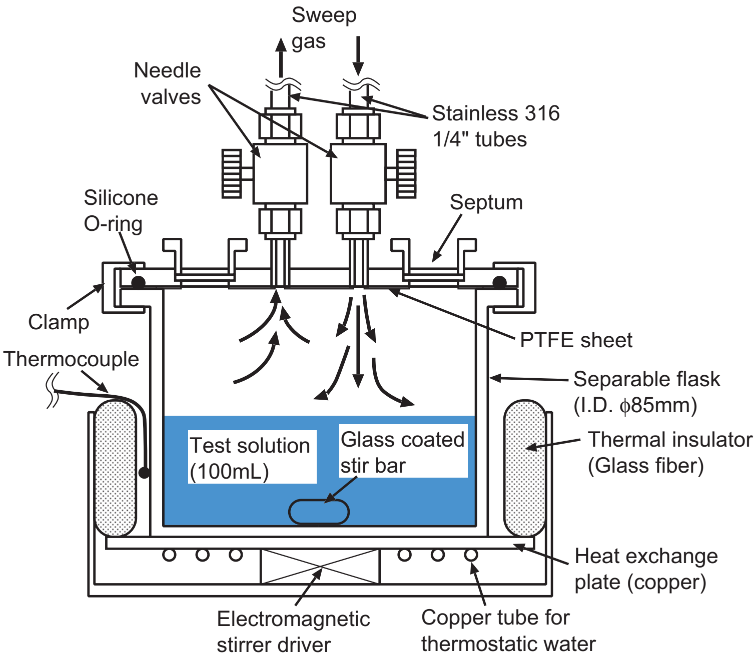

Figure 2.3: Irradiation vessel.

2.1.3 Irradiation vessel

Figure 2.3 schematically shows the vessel for irradiation of the test solution. Figure 2.4 shows the set-up of the vessel in the irradiation chamber. A separable flask of 300 mL nominal capacity and 85 mm inner diameter was covered by a specially designed lid of electro-polished 316 stainless steel. The liquid and gas volumes in the vessel were 100mL and 286mL, respectively. The inner wall of the lid was covered by 0.2 mm thick PTFE sheet to avoid iodine adsorption on the surface. The lid had two access holes of 17 mm diameter covered by septa. The lid had two 1/4" diameter nozzles to which needle valves (SS-14DPS4, Swedge Lock) for the inlet and outlet were attached. The solution was stirred with a glass coated 6 mm diameter and 25 mm long stir bar driven by electromagnetic stirrer driver (Remote Micro type, VARIOMAG). The temperature of the vessel was controlled at 25±2 °C during irradiation by providing thermostated water to the copper tube coil soldered under the copper plate on which the vessel was set.[1]

The gas volume in the vessel was swept at a constant flow rate, 1 L/min, corresponding to the replacement rate about 3.5 min^{-1}. The tube between the vessel and the iodine sampler had a volume 74 cm^3 (4.35 mm inner diameter and 5m long), which was replaced in about 4.5 s.

2.1.4 Iodine Sampling and On-line Counting

Figure 2.5 shows the iodine samplers. Two kinds of filter elements were used in separate samplers and those were set in tandem in the iodine sampling system as shown in Fig. 2.1, and in detail in Fig. 2.6. The first sampler contained 5 pieces of Ag coated (0.3mg/cm^2 by physical vapor deposition (PVD)) glass fiber filter elements (No.50, Toyo Roshi) that selectively collects molecular iodine. The second sampler contained 3 pieces of $AgNO_3$ impregnated alumina particle bed filter elements that collects both molecular iodine and organic iodine. Connected in that way, the first sampler collects molecular iodine and the second collects organic iodine. This species separating iodine sampling system had been developed and proved at former Japan Atomic Energy Research Institute (JAERI).(Appendix of Ref. [11])

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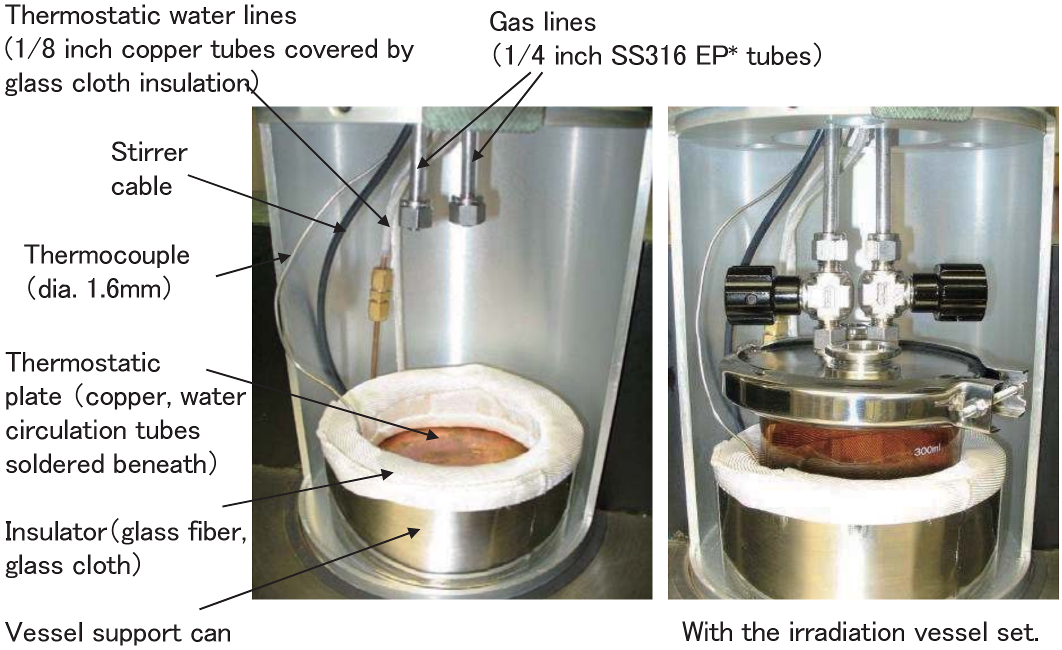
[1] This temperature control was not provided in early runs, namely 19-01–03 in Table 2.1.



With the irradiation vessel set.

* stainless steel 316, electropolished

Figure 2.4: Set-up of the irradiation vessel in the irradiator.

A diffuser ring made of PTFE was inserted at the inlet of each sampler so that the first filter element was effectively exposed to the gas flow. The iodine samplers were connected in the flow system by quick couplers (Semicon Cupla SP 316 stainless type or PTFE type, Nitto Kohki) that have built-in valve mechanism to automatically shut the flow path when disconnected.

The samplers were γ-counted by NaI(Tl) scintillators (4” hole type by SIONIX with instrumentation by Seiko EG&E). The γ-counting for the two samplers was continued through out a run and the cumulative counts in the energy range corresponding to 364 keV γ-ray of ^{131}I were read out in every 5 minutes. The instantaneous activity in a sampler was, then, calculated from the difference of two successive readouts. The statistical uncertainty in the activity in this method corresponded to $\sim$0.05% of the total initial iodine inventory in the present experiments.

The geometric efficiency of the on-line γ-counting depended on the distribution of iodine deposition in the sampler and needed calibration for every run. It was done by analyzing each filter element—whose geometric efficiency was known—after a run by another off-line NaI counter and by comparing the on-line and the off-line results. The off-line measurement was more precise, *i.e.* the relative statistical uncertainties for each sector: aqueous, Ag filters and $AgNO_3$ filters, were $< 0.5\%$, $< 1\%$ and $\sim$10%, respectively, even if their fractions to the total initial inventory was less than 0.1%. The mass balance of iodine in each run was checked with the off-line data.

The samplers as well as NaI scintillators were confined in a chamber shielding them by 100 mm thick lead from a high background radiation due to the γ-irradiator. We kept the temperature inside the chamber at about 20 °C by heaters and insulators not to have condensation of vapor in the samplers. Higher temperatures were not recommended for assurance of correct functioning of the activity instrumentation, though preferable for prevention of condensation.

Figure 2.5: Iodine samplers.

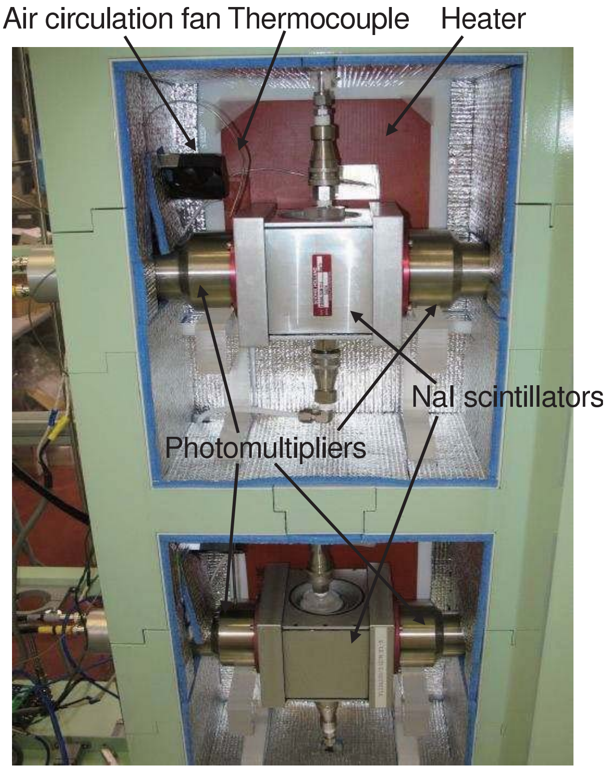


Figure 2.6: On-line counting chamber.

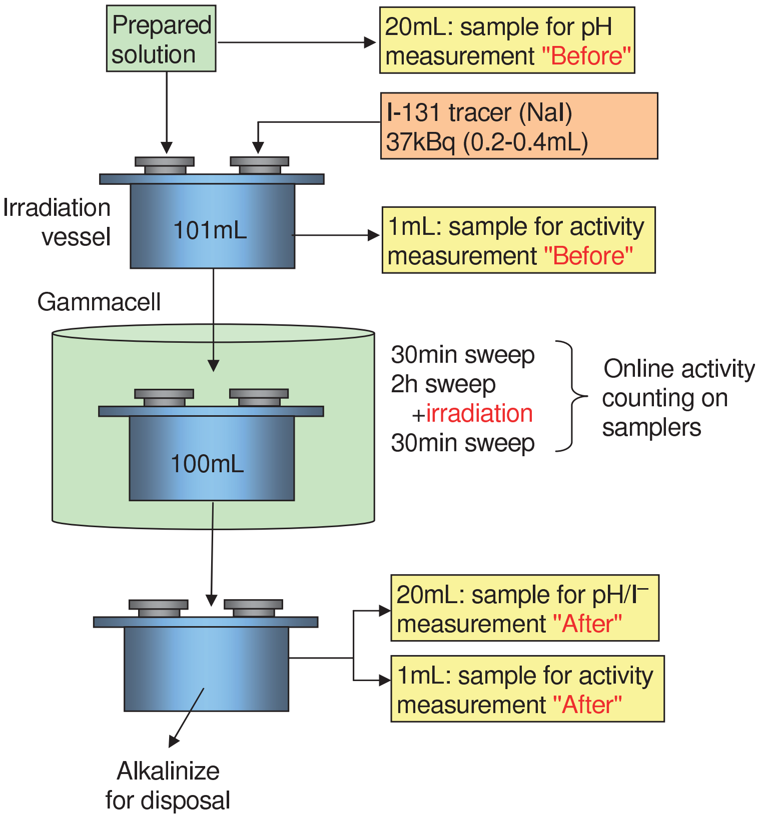


Figure 2.7: Experimental procedure.

2.2 Procedure

Figure 2.7 shows the procedure of the experiment. The prepared test solution 121 mL was put in the test vessel, and 20 mL was taken for the initial I^- and pH measurement. Then, ^{131}I tracer was added about 37 kBq (volume 5–200μL) and 1 mL was sampled for the initial activity measurement.

The irradiation vessel, now containing 100mL of the test solution, was set in the γ-irradiator and the gas phase was swept with prescribed gas for 30 minutes, during which the solution became saturated with the gas. The solution was kept stirred at 350 rpm. No significant disturbance in the gas-liquid interface was caused by the stir. Two hours of irradiation and 30 minutes of no-irradiation period followed. The gas sweep and stir were continued through out the process.

The vessel was removed from the irradiator and the solution was sampled for the measurement of remaining activity, I^- concentration and pH.

Measurement of pH and I^- concentration was done by a pH electrode (8102, Thermo) and an ion selective electrode (ISE) (9653, Thermo).

2.3 Conditions and Parameters

The experimental conditions and variation of the parameters are summarized in Table 2.1. The air swept case with 1×10^{-4} M CsI solution without additives except the pH buffer at temperature 25 °C was taken as the "base case". The gas flow rate and the stir revolution rate that determine the interface mass transfer were kept constant, 1 L/min and 350 rpm, respectively.

The interface mass transfer coefficient and its dependence on the stir, gas flow rate, tem-

Table 2.1: Experimental conditions: gaseous iodine release experiments

Name	No.	Irradiation		Iodine initial conc.(M)	Temper - ature (°C)	Stir rate (rpm)	Sweep gas	Initial pH	Additives (ions,organics,etc.) conc. (M)	Parameters, etc.	Notes
		Dose rate (kGy/h)	Irrad. time (h)								
	19-01	7.9	2	1.0E-04	30-40	350	Air	5.36		pH=5.5	
	19-02	7.7	2	1.0E-04	30-40	350	Air	6.75		pH=7	
	19-03	7.7	2	1.0E-04	30-40	350	Air	8.41		pH=9 (intended)	Contamination of the line due to the previous case
pH9	19-03r	7.5	2	1.0E-04	20-28	350	Air	9.10		pH=9	
N2	19-04r	7.5	2	1.0E-04	24-27	350	N2	6.72		N2	
pH9-N2	19-05	7.6	4	1.0E-04	(25-35)	350	N2	9.13		pH=9, N2	Lack of temperature data
Fe	19-06m	7.5	2	1.0E-04	22-25	350	Air	6.85	Fe2+ 1E-4	Fe ions	
OrgL	19-07m	7.4	2	1.0E-04	21-27	350	Air	6.99	MIBK 1E-4	Organic additive	
OrgL-N2	19-08m	7.4	2	1.0E-04	23-26	350	N2	6.99	MIBK 1E-4	Organic additive, N2	
Cl	19-09	7.2	2	1.0E-04	20-26	350	Air	7.03	Cl- 1E-3	Cl- ions	
OrgH	20-01	6.9	2	1.0E-04	25-27	350	Air	6.93	MIBK 1E-3	Organic additive (high)	
OrgH-N2	20-02	6.9	2	1.0E-04	25-29	350	N2	7.16	MIBK 1E-3	Organic additive (high), N2	
OrgH-Ar	20-03	6.9	2	1.0E-04	24-26	350	Ar	6.98	MIBK 1E-3	Organic additive (high), Ar	
T40	20-04	6.8	2	1.0E-04	38-39	350	Air	7.01		Temperature	
T40-pH5.5	20-05	6.7	2	1.0E-04	39-40	350	Air	5.60		Temperature, pH=5.5	
OrgH-LO	20-06	6.7	2	1.0E-04	25-26	350	N2-2%O2	7.04	MIBK 1E-3	Organic additive (high), Low O2	
LO	20-07	6.6	2	1.0E-04	25-26	350	N2-2%O2	7.03		Low O2	
Base	20-08	6.6	2	1.0E-04	24-26	350	Air	6.97			
pH5.5	20-09	6.5	2	1.0E-04	25-26	350	Air	5.55		pH=5.5	
OrgL-LO	20-10	6.5	2	1.0E-04	24-26	350	N2-2%O2	7.12	MIBK 1E-4	Organic additive, Low O2	
ClH	21-01	6.1	2	1.0E-04	24-26	350	Air	7.06	Cl- 1E-2	Cl- ions	
ClL	21-02	6.1	2	1.0E-04	24-26	350	Air	6.97	Cl- 1E-4	Cl- ions	
H2	21-03r	5.8	2	1.0E-04	24-26	350	N2-5%H2	7.08		N2-5%H2	
N2a	21-04r	5.8	2	1.0E-04	25-26	350	N2	7.07		N2	
H2Ar	22-01	5.3	2	1.0E-04	25	350	Ar-5%H2	7.01		Ar-5%H2	
Ar	22-02	5.3	2	1.0E-04	25	350	Ar	6.98		Ar	
H2LAr	22-03	5.3	2	1.0E-04	25	350	Ar-3%H2	6.99		Ar-3%H2	
OrgL-MO	22-04	5.3	2	1.0E-04	25	350	N2-5%O2	6.99	MIBK 1E-4	Organic additive, Low O2 (5%)	
MO	22-05	5.3	2	1.0E-04	25	350	N2-5%O2	7.01		Low O2 (5%)	

Common condition	Initial iodine speciation=CsI, Pressure=0.1MPa, Liquid volume=100mL, Gas volume=286mL, Gas-liq. interface area=0.567dm2(ϕ85mm), Gas flow rate=1L/min, Stirrer=ϕ6mm x 25mm glass. I-131 tracer added (about 37kBq), pH buffer=0.1M B(OH)3-NaOH

perature and species was separately examined by a complementary experiments described in Appendix A. The overall interface mass transfer coefficient in the standard condition, *i.e.* 25 °C and 1 L/min gas flow with 350 rpm stir, was 4×10^{-4} dm/s for oxygen and 1×10^{-4} dm/s for molecular iodine. The coefficients for other species depend on their molecular volumes, and are evaluated by Eq.(A.11) in Appendix A.

In the following, the parameters we examined, their ranges in the experiment, and the relevance to the prototypic condition are described.

Iodine concentration The inventory of iodine accumulates in a 1 GWe class reactor core is about 80 mol after several years of operation with a fuel replacement for 1/4–1/3 of the core every year. The maximum concentration of iodine in the water phase of a containment vessel is deduced from that. The amount of water in the PWR sump or BWR supression chamber is in the range 2000–4000 m^3. With an assumption that iodine is dissolved in the water as I^-, it gives concentrations in the order of 10^{-5} M. In the present experiments, as a maximum possible value, we took 1×10^{-4} M. The influence of the initial iodide concentration was examined by previous works[7, 9], and we did not change it in our work.

pH It is known from previous studies[7, 9, 3] that pH has the most significant influence on iodine volatility, low pHs enhance volatility and high pHs lower it. The factors that affects the pH in the CV of LWRs were reviewed by Beahm et al.(1992)[12]. The range of pH differs by plant types. If boric acid for reactivity suppression and an alkalinizer for iodine control are put in, they control the pH. Other factors include alkaline FPs such as CsOH and $CsBO_3$, HNO_3 by humid air radiolysis, CO_2 by decomposition of organic impurities or concrete, HCl by degradation of chlorine containing organic compounds such as cable insulations. If the pH is not intentionally controlled, the incoming alkaline FPs raise the pH to 9–10, and other factors decrease it. The extent of the pH drop has a large uncertainty depending on the inventory of the materials concerned.

Though the influence of pH had already been examined comprehensively, we studied it again considering the importance and to check the consistency of our results with existing data. We controlled the pH in the range 5.5–9 with 0.1 M boric acid-sodium hydroxide buffers.

Temperature Ashmore et al.(2000)[7] showed experimentally the influence of temperature on the inorganic iodine volatility. Their data showed decreased volatility when the temperature was raised from 25 °C to 40–70 °C. The volatility decrease is mechanistically explained by the enhancement of iodine hydrolysis at high temperatures. An opposite effect is done by the temperature dependence of the gas-aqueous partition coefficient of iodine, that gives more volatility at high temperatures.[13, 14, 15] Those two effects compete; either of which prevails probably depends on pH. So, we examined the influence of temperature, 25 °C and 40 °C, at two pHs, 5.5 and 7.

Iron ions Mechanistic models include a catalytic reactions of Fe^{2+} and Fe^{3+} ions that converts $\cdot O_2^-$, a primary reductant of iodine, to O_2 and H_2O_2.[14, 13, 15] It is known that such reactions well explain some data from experiments with steel vessels[16, 15], especially about the iodine volatility at high pHs > 7, *i.e.* models under predict the volatility without those reactions. We examined the effect of iron ions by putting in 1×10^{-4} M of Fe^{2+} (as $FeSO_4$), for it had not been confirmed experimentally.

HCl The amount of HCl by degradation of chlorine containing organic compounds such as cable insulations may be quite large, corresponding to concentrations in water in CVs up to the order of 10^{-3} M.[12, 17, 18] The review by Beahm et al.[12] showed the inventories in several plant types ranging 4300–36000 mol, giving concentrations 10^{-3}–10^{-2} M with a typical water volume 4000 m^3. A part of its influence is the impact on pH, and another is a possible interference with reactions of iodine species, because they are in the same elemental group. We looked into the latter point and added Cl^- ions (as NaCl) up to 1×10^{-2} M.

Oxygen In irradiated aqueous solutions, dissolved oxygen produces a primary reductant of iodine, $\cdot O_2^-$. Thus, the absence of oxygen enhances the oxidation from I^- to volatile I_2 and increases the iodine volatility.[14, 13, 15] This effect was experimentally observed by Ashmore et al.[7]. It is important in terms of the conditions in BWR CVs that are purged of oxygen. We examined the effect of oxygen by changing the sweep gas between air and nitrogen or argon, and also looked into its coupling with the influence of organic compounds, as described in the next paragraph.

Organic compound Organic impurities in the solution cause production of organic iodines, some of which are more volatile than elemental iodine and less adsorptive on walls, thus more persistently air borne. Also, the decomposition of organics in irradiated aqueous solution produces acids that decrease pH. A possible major source of such organic impurities is the organic solvents remaining in the paint on the containment wall[2, 3], typically, ketones such as acetone, methylethylketone, methylisobutylketone, and aromatics such as toluene, xylene, ethylbenzene. Such organic iodine production was previously investigated in middle scale and small scale experiments[19, 20, 8], mostly in aerated environment assuming PWR or CANDU plants. We lacked data at low oxygen concentration conditions relevant to BWR CVs.

We examined the influence of organic compounds combined with the influence of oxygen systematically. We used MIBK as one of the organic solvents known to be used in paints applied in CVs of LWRs, and also changed the oxygen concentration in the sweep gas: air (21%), low oxygen concentrations relevant to BWRs (5% and 2%), and nitrogen or argon (0%).

Hydrogen During the progression of a severe accident of LWRs, the zircaloy in the fuel clad is likely to be oxidized and it produces a large amount of hydrogen. In BWRs, the CV atmosphere is purged of oxygen with nitrogen so that the hydrogen does not burn. The partial pressure of hydrogen in the CV rises up to 0.1 MPa, comparable to the initially existing nitrogen and about 50% of the total pressure[21]. Ashmore et al.[7] examined the influence of hydrogen with air containing 4% H_2 and observed decreased iodine release rate. We examined it by adding hydrogen to the sweep gas by up to 5% concentrations. The rest of the gas was an inert gas, either nitrogen or argon.

The runs were named according to parameter settings as indicated in Table 2.1. The cases for comparison are summarized by the category as follows.

pH : Base, pH5.5, pH9

Temperature : Base, T40, pH5.5, T40-pH5.5

Fe^{2+} ion : Base, Fe

Cl^- ion : Base, Cl, ClL, ClH

O_2/Organic : Base, OrgL, OrgH, MO, OrgL-MO, LO, OrgL-LO, OrgL-N2, N2a, OrgL-N2, OrgH-N2, OrgH-Ar

H_2 : N2a, H2, Ar, H2LAr, H2Ar

Those names are used to call the runs hereafter.

The reagents used in the experiment were as follows.

- Cesium iodide: 99.9%, Wako Pure Chemical Industries
- Boric acid: >99.5% ACS reagent grade, Sigma-Aldrich
- Sodium hydroxide: >96%, Wako Pure Chemical Industries
- Sodium chloride: >99%, ACS reagent grade, Sigma-Aldrich
- Ferrous sulfate: >99.5%, Kanto Chemicals
- Methylisobutylketone (MIBK): >99.5%, Wako Pure Chemical Industries
- o-Xylene: >98%, Wako Pure Chemical Industries
- ^{131}I as tracer: Perkin Elmer NEZ035H 74 MBq (>185 GBq/mg (as iodine) in 0.1 mol/L NaOH aq. (pH=12–14)) diluted into pure water (∼1.8 MBq/mL)
- Water: pure water supplied by Millipore Milli-Q Academic A10, >18.2 MΩm@25 °C, <40 ppb TOC (Total Organic Carbon)

The reagents were used without further purification.

2.4 Results and Discussions

2.4.1 Mass Balance

Figure 2.8 shows the mass balance of iodine. For every case, the stacked columns indicate, from the bottom, the fractions in the test solution, in the filters at the end of the run, and the loss calculated by subtracting the former two from 100%. It is observed that the loss becomes larger when larger fraction was released. The loss was usually a few percents, and at most ∼10% in the cases of acidic conditions. The present experiment showed a good mass preservation.

A primary cause of the loss is probably the adsorption in the tube connector at the entrance of the first iodine sampler where corresponding order of activity was detected by the off-line counting.

We assumed the lost fraction was released as I_2 that is much more adsorptive than organic iodines. In the following, “total released” or “released as I_2” means the sum of the fraction collected by Ag filters and the loss.

2.4.2 Iodine Release Behavior and Parametric Effects

Table 2.2 summarizes the experimental results. Figures 2.9–2.20 show the comparison of release histories and the final release fractions in terms of the parametric effects. Detailed descriptions of each run and individual data plots are provided in Appendix B.

pH Figure 2.9 shows the histories of total iodine release for the cases of various pHs. The figure (a) shows the release history along the absorbed dose, while the figure (b) shows that along time. Generally in the present series of experiments, the release was observed during the irradiation and the release was nearly linear in terms of time.

Figure 2.10 compares the final release fractions, including early cases in which the temperature control was not implemented yet. The comparison shows that the total release fraction increased more than 2 orders for the pH change from 9 to 5.5. It confirms that pH is the dominant factor for iodine volatility and it is consistent with existing data[7, 9].

Table 2.2: Summary of the experimental results: gaseous iodine release experiments

Name	No.	Initial pH	Final pH	Initial I- conc.(M)	Final I- conc.(M)	Final iodine frac. in solution (%)	Final I- frac. in solution (%)	Filters total frac. (%)	AgNO3 filter frac. (%)	Loss (%)	Iodine release frac. (%)	AgNO3/ Release (%)
	19-01	5.36	5.63	1.00E-4	2.62E-5	31.0	26.2	57.7	0.046	11.27	69.0	0.066
	19-02	6.75	6.67	1.00E-4	7.97E-5	87.9	79.7	9.48	0.069	2.61	12.1	0.57
	19-03	8.41	8.39	1.00E-4	8.87E-5	97.6	88.7	0.79	0.103	1.59	2.38	4.3
pH9	19-03r	9.10	9.09	1.00E-4	-	104	-	0.25	0.022	-	0.25	8.74
N2	19-04r	6.72	6.77	1.00E-4	5.01E-5	53.1	50.1	40.8	0.272	6.08	46.9	0.58
pH9-N2	19-05	9.13	9.13	1.00E-4	6.99E-5	97.4	69.9	1.12	0.097	1.48	2.60	3.71
Fe	19-06m	6.85	6.73	1.00E-4	8.11E-5	86.7	81.1	11.1	0.024	2.25	13.3	0.18
OrgL	19-07m	6.99	6.92	1.00E-4	7.98E-5	91.3	79.8	8.41	0.056	0.33	8.74	0.64
OrgL-N2	19-08m	6.99	6.96	1.00E-4	6.52E-5	80.9	65.2	12.3	0.544	6.86	19.1	2.85
Cl	19-09	7.03	7.00	1.00E-4	9.30E-5	89.9	-	10.8	0.061	-	10.8	0.57
OrgH	20-01	6.93	6.37	1.00E-4	8.06E-5	86.5	80.6	9.74	0.408	3.74	13.5	3.03
OrgH-N2	20-02	7.16	7.10	1.00E-4	8.38E-5	97.7	83.8	2.18	0.816	0.14	2.32	35.2
OrgH-Ar	20-03	6.98	6.96	1.00E-4	8.78E-5	98.0	87.8	1.66	0.542	0.36	2.02	26.8
T40	20-04	7.01	6.98	1.00E-4	9.34E-5	93.0	93.4	4.66	0.039	2.33	6.99	0.56
T40-pH5.5	20-05	5.60	5.66	1.00E-4	3.94E-5	40.3	39.4	54.0	0.086	5.74	59.7	0.14
OrgH-LO	20-06	7.04	6.96	1.00E-4	7.70E-5	93.4	77.0	4.70	1.52	1.87	6.57	23.1
LO	20-07	7.03	7.02	1.00E-4	8.07E-5	85.8	80.7	12.0	0.029	2.24	14.2	0.20
Base	20-08	6.97	6.91	1.00E-4	8.32E-5	90.5	83.2	7.66	0.018	1.85	9.51	0.19
pH5.5	20-09	5.55	5.81	1.00E-4	1.87E-5	21.6	18.7	70.6	0.096	7.76	78.4	0.12
OrgL-LO	20-10	7.12	7.03	1.00E-4	8.68E-5	89.0	86.8	11.2	0.166	-	11.2	1.48
ClH	21-01	7.06	-	1.00E-4	8.78E-5	91.2	87.8	9.17	0.016	-	9.17	0.17
ClL	21-02	6.97	6.93	1.00E-4	8.82E-5	90.6	88.2	8.06	0.018	1.39	9.45	0.19
H2	21-03r	7.08	7.07	1.00E-4	7.73E-5	85.0	77.3	13.9	0.020	1.09	15.0	0.13
N2a	21-04r	7.07	7.06	1.00E-4	7.35E-5	75.0	73.5	21.8	0.059	3.15	25.0	0.24
H2Ar	22-01	7.01	7.03	1.00E-4	8.77E-5	87.6	87.7	10.9	0.017	1.45	12.4	0.14
Ar	22-02	6.98	7.04	1.00E-4	7.36E-5	79.4	73.6	17.6	0.027	3.02	20.6	0.13
H2LAr	22-03	6.99	7.01	1.00E-4	9.02E-5	84.3	90.2	13.5	0.027	2.26	15.8	0.17
OrgL-MO	22-04	6.99	6.97	1.00E-4	7.94E-5	88.8	79.4	9.37	0.071	1.84	11.2	0.64
MO	22-05	7.01	7.02	1.00E-4	7.95E-5	90.0	79.5	8.76	0.018	1.26	10.0	0.18

Temperature Figures 2.11 and 2.12 show the histories of total iodine release and comparison of the final release fractions, respectively, for the cases of different temperatures at pHs 7 and 5.5. Between the temperatures 25 and 40 °C, the higher temperature made lower release fractions at both pHs. In the present range of conditions, the effect of I_2 hydrolysis decreasing the gaseous I_2 release prevailed over a possible opposite effect of the temperature dependence of I_2 partition coefficient. The observed decrease of iodine volatility at higher temperatures is consistent with a previous study[7].

Inorganic impurities Figure 2.13 shows the histories of total iodine release for the cases Fe^{2+} or Cl^- ions were added. The results showed no significant influence of those ions.

Despite that mechanistic models showed a significant influence of the catalytic reaction of iron ions[13, 14, 15], it was experimentally not observed. Other researchers[7] also reported a result similar to the present experiment.

Organic impurities and oxygen Fugures 2.14 and 2.15 show the histories of total iodine release for the cases with various MIBK concentrations (aqueous) and oxygen concentrations (sweep gas). Figures 2.16 and 2.17 show the histories of organic iodine release. Figure 2.18 shows the comparison of final release fractions as I_2 (a) and organic iodine (b).

Comparison of the total or I_2 release fractions in the cases without MIBK addition (Base, MO, LO and N2a) showed a trend that lower oxygen concentrations caused higher release fractions. It was consistent with predictions by mechanistic models[14, 13, 15] and a previous work[7]. When MIBK was added, the I_2 release fraction decreased with increasing MIBK concentrations in low or no oxygen concentration cases (-LO and -N2). The organic iodine release fraction increased with increasing MIBK concentrations with all the oxygen concentrations. However, the relation between the organic iodine release fractions and the oxygen concentration was not straight at high MIBK concentration (Fig. 2.18 (b), MIBK 10^{-3} M cases). This rather complicated dependence is explained by competition on the oxygen consumption between iodine and organic reactions. Detailed discussions are given elsewhere[11, 15].

Hydrogen Figures 2.19 and 2.20 show the histories of total iodine release and comparison of the final release fractions, respectively, for the cases with hydrogen containing sweep gases. For both the nitrogen based and argon based hydrogen-containing gases, decrease of the total iodine release fractions were observed with increasing hydrogen fractions. Both showed about 40% decrease at 5% hydrogen.

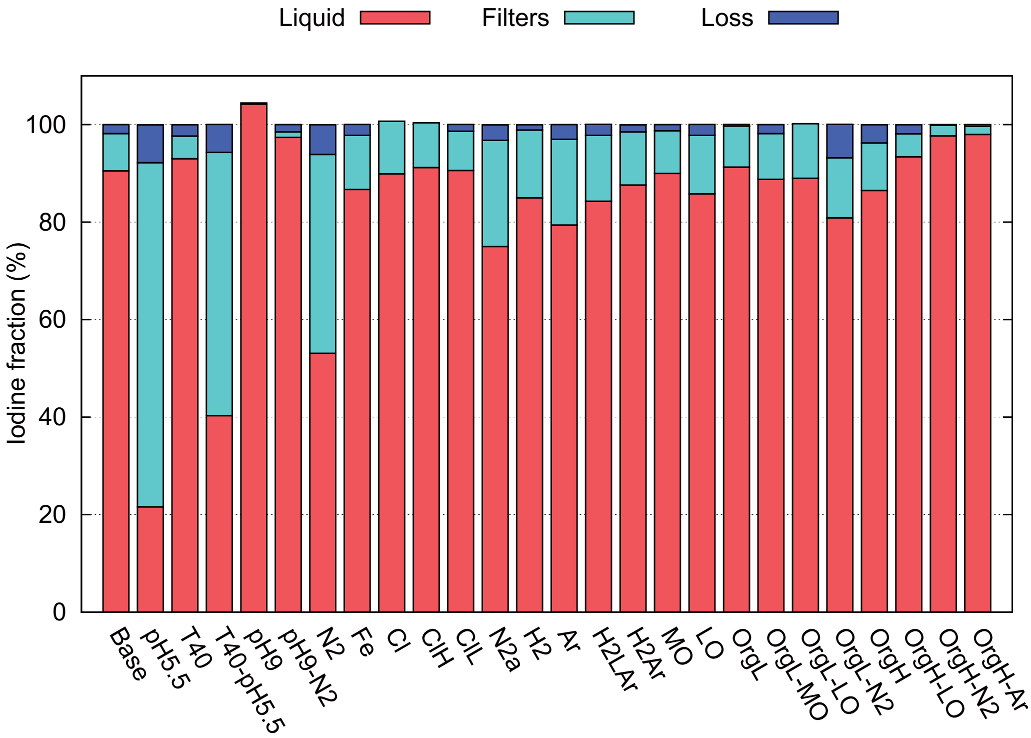
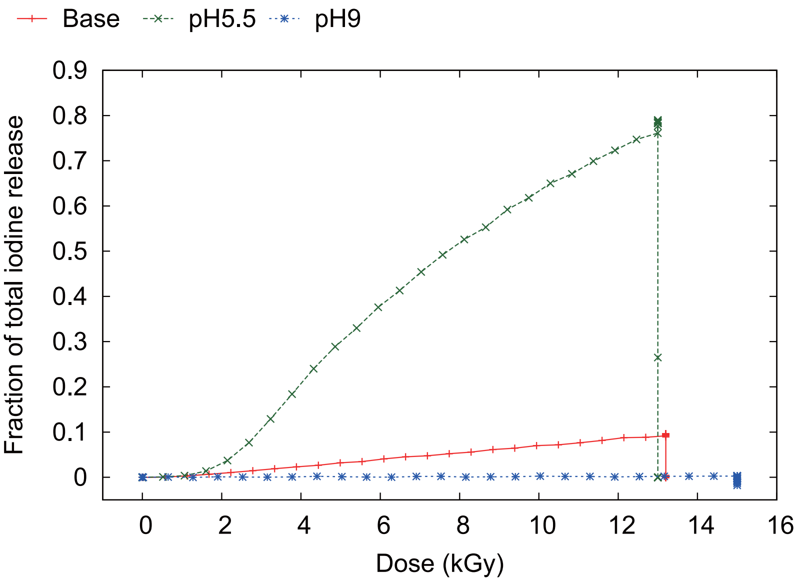
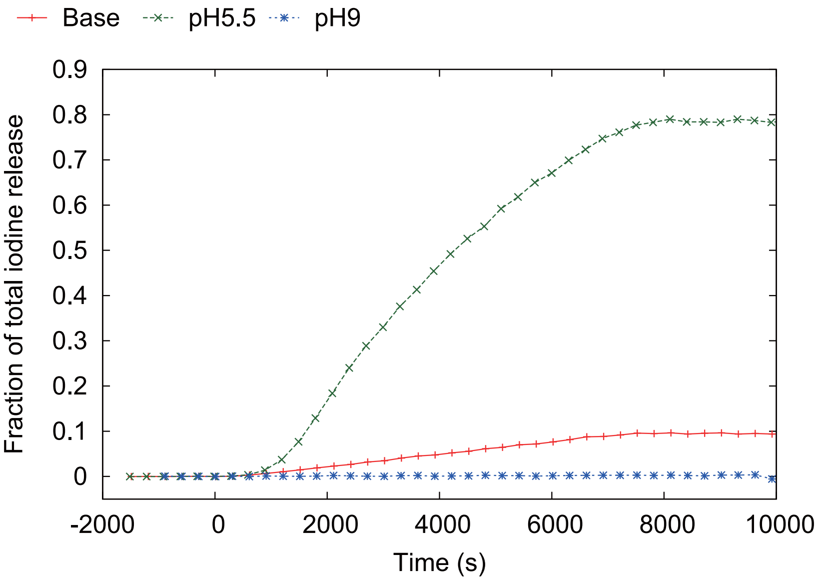


Figure 2.8: Mass balance of iodine in the present experiment.



(a) Evolution of release in terms of absorbed dose.



(b) Evolution of release along time.

Figure 2.9: Evolution of total gaseous iodine release: pH dependence.

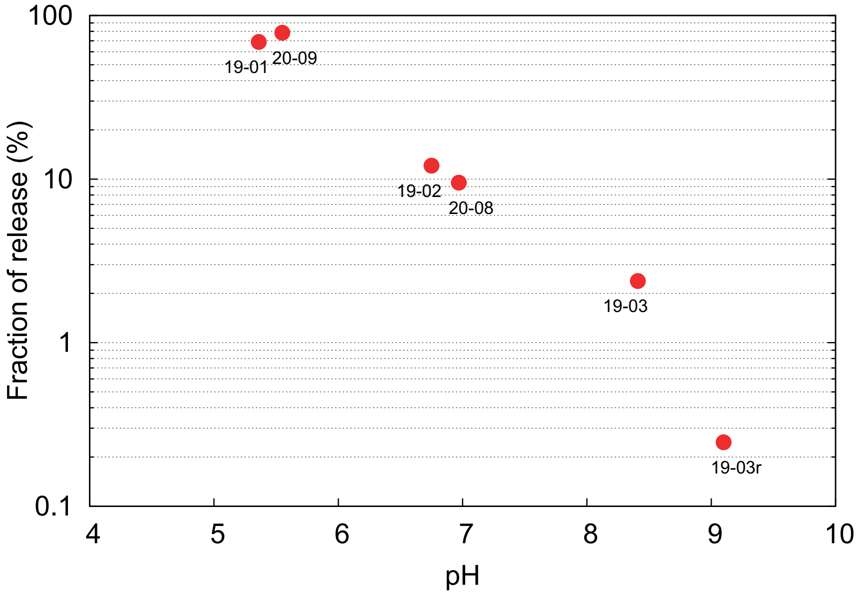
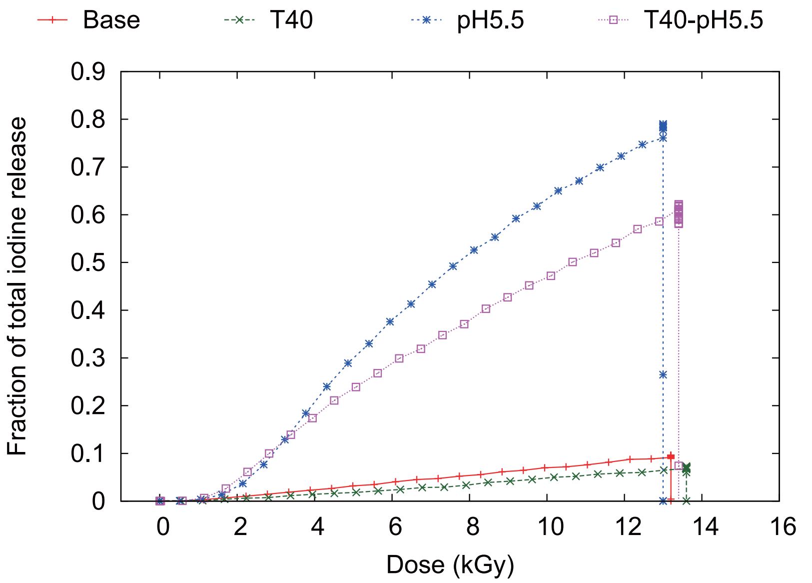
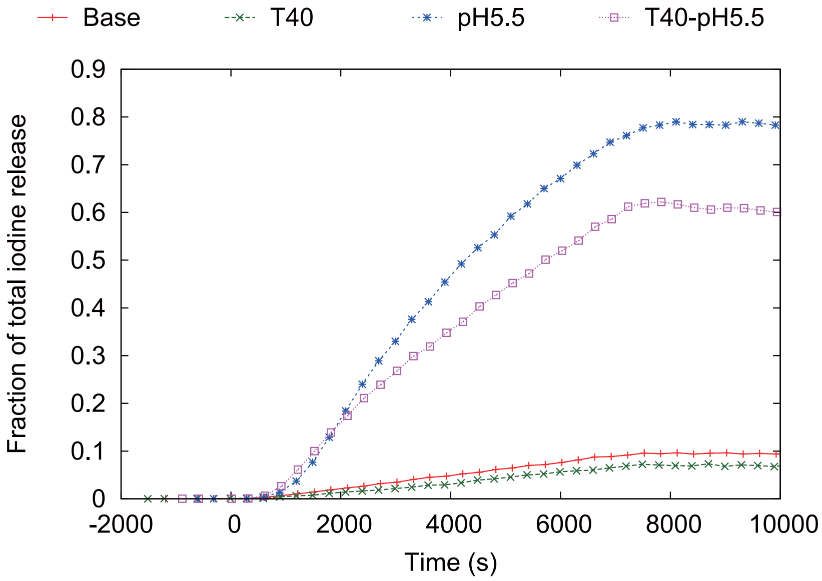


Figure 2.10: Comparison of the final release fractions (total iodine): pH dependence.



(a) Evolution of release in terms of absorbed dose.



(b) Evolution of release along time.

Figure 2.11: Evolution of total gaseous iodine release: temperature dependence at pH=7 and 5.5.

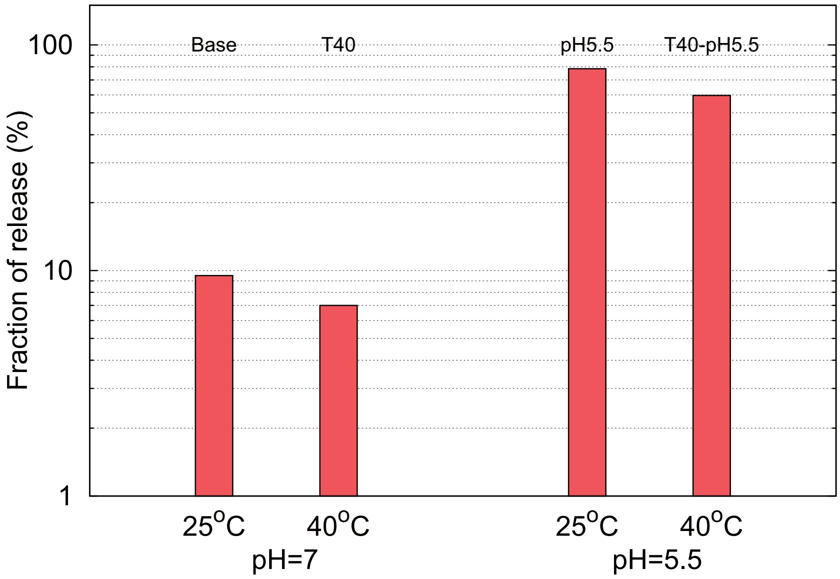
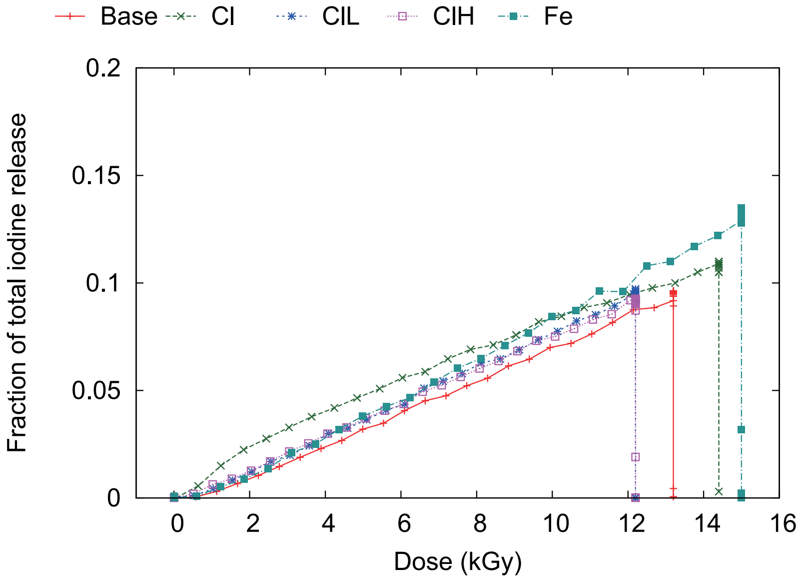
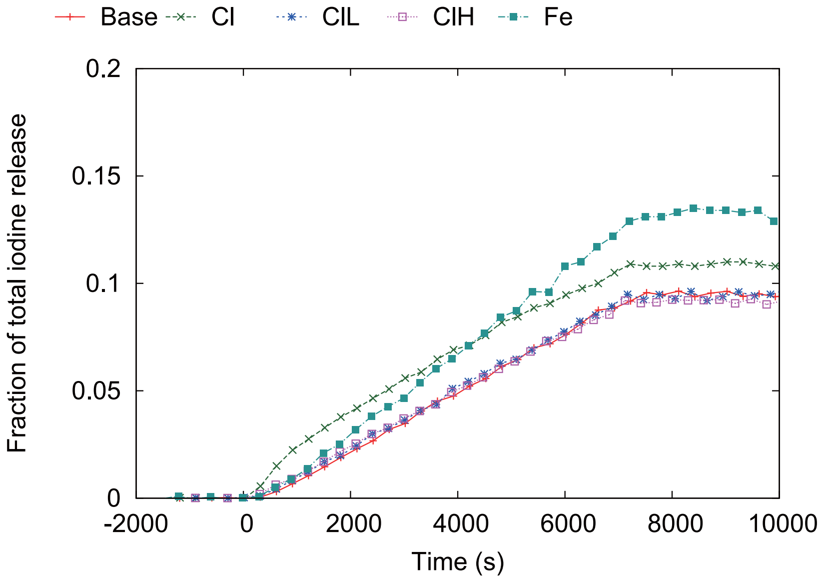


Figure 2.12: Comparison of the final release fractions (total iodine): influence of temperature at pH=7 and 5.5.

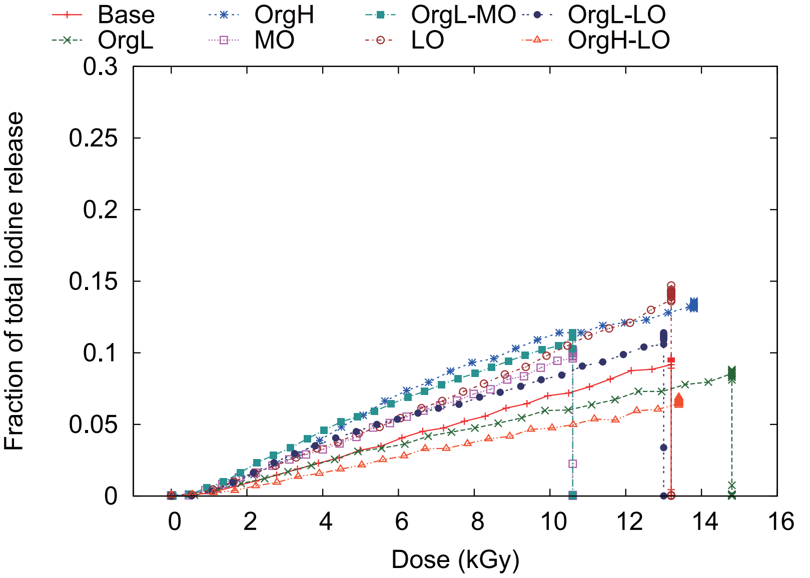


(a) Evolution of release in terms of absorbed dose.

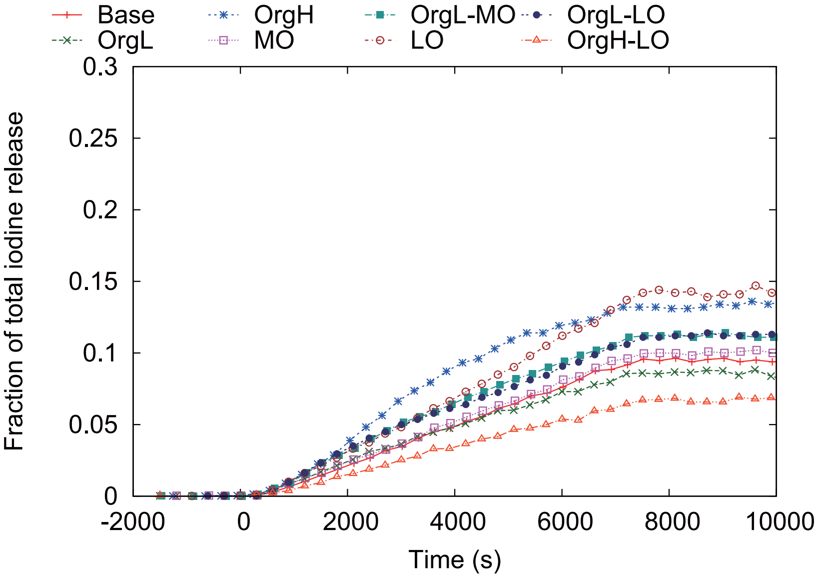


(b) Evolution of release along time.

Figure 2.13: Evolution of total gaseous iodine release: influence of inorganic impurities (chloride and iron ions).

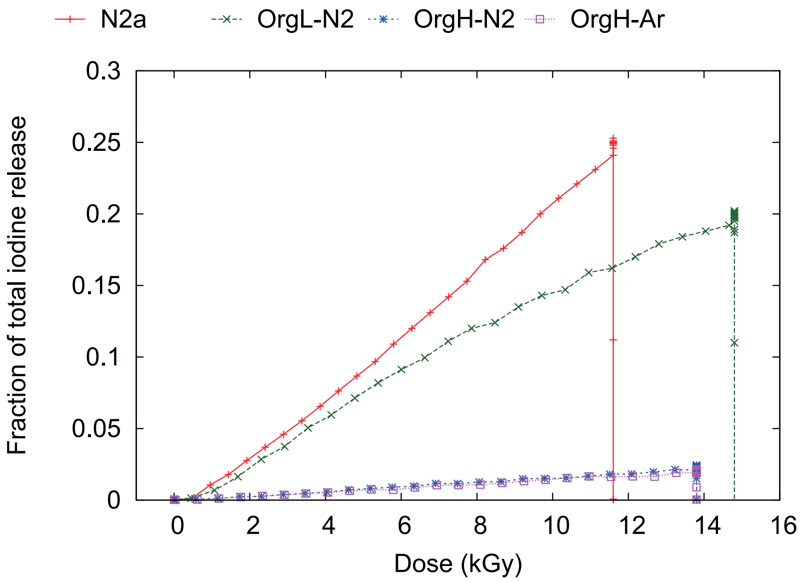


(a) Evolution of release in terms of absorbed dose.

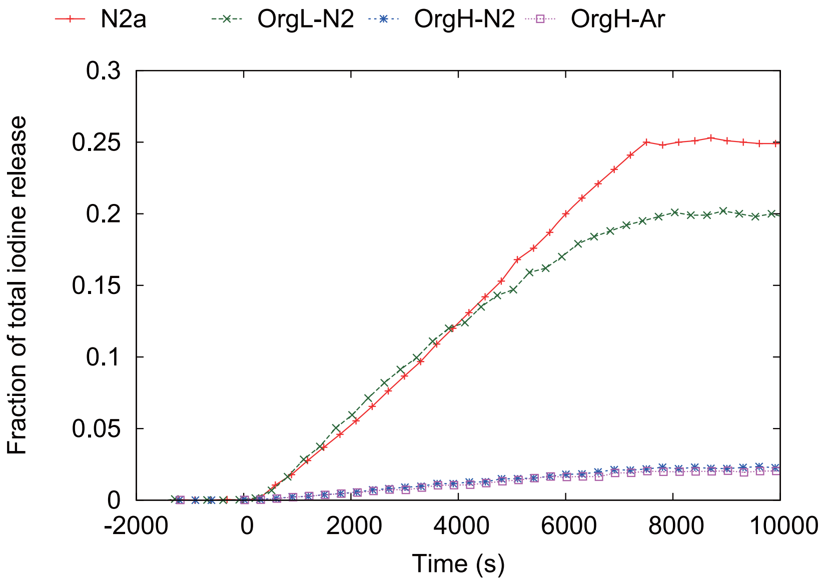


(b) Evolution of release along time.

Figure 2.14: Evolution of total gaseous iodine release: influence of organic impurity (MIBK) and oxygen concentration. (air and low oxygen conditions)

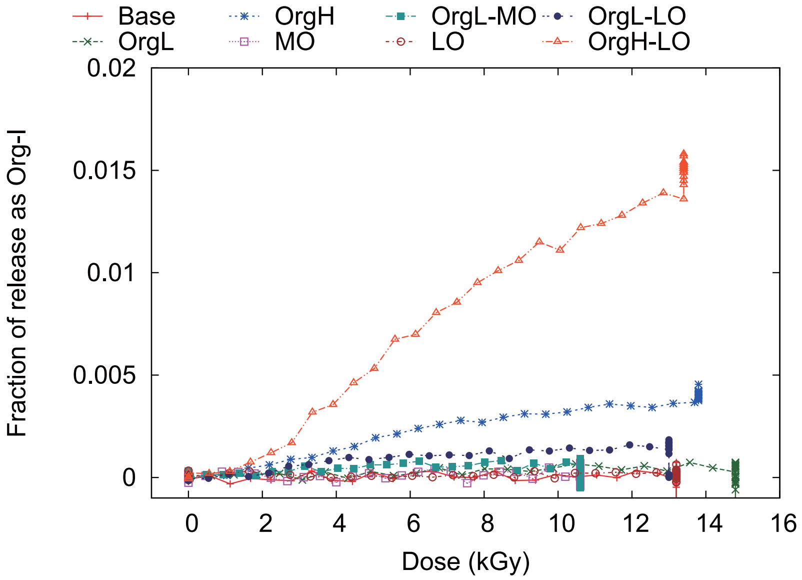


(a) Evolution of release in terms of absorbed dose.

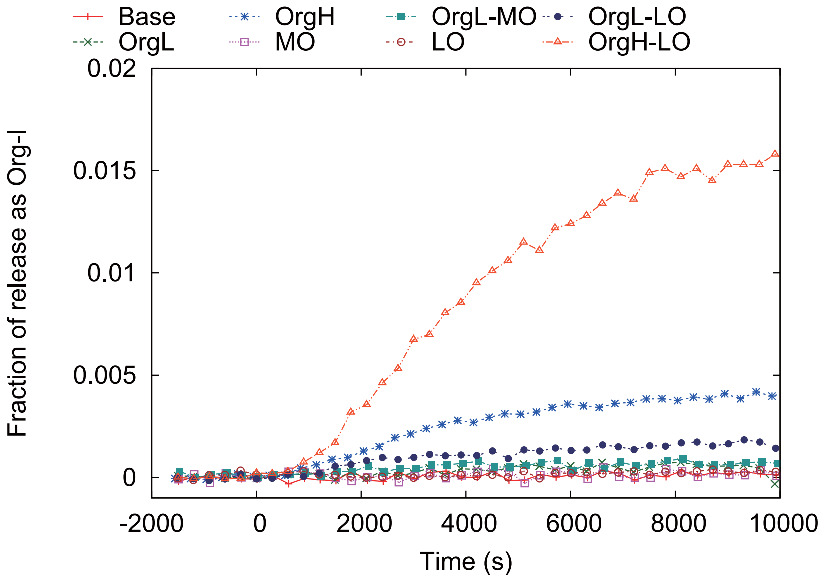


(b) Evolution of release along time.

Figure 2.15: Evolution of total gaseous iodine release: influence of organic impurity (MIBK) and oxygen concentration. (no oxygen conditions)

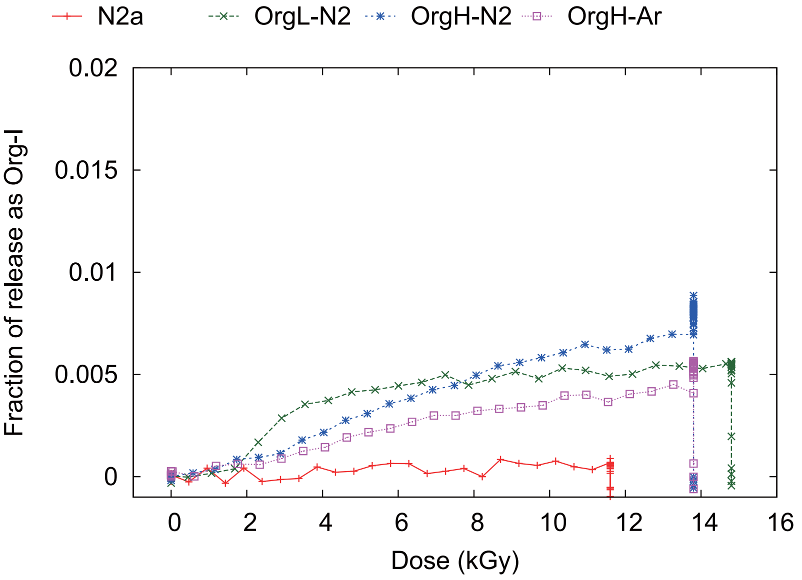


(a) Evolution of release in terms of absorbed dose.

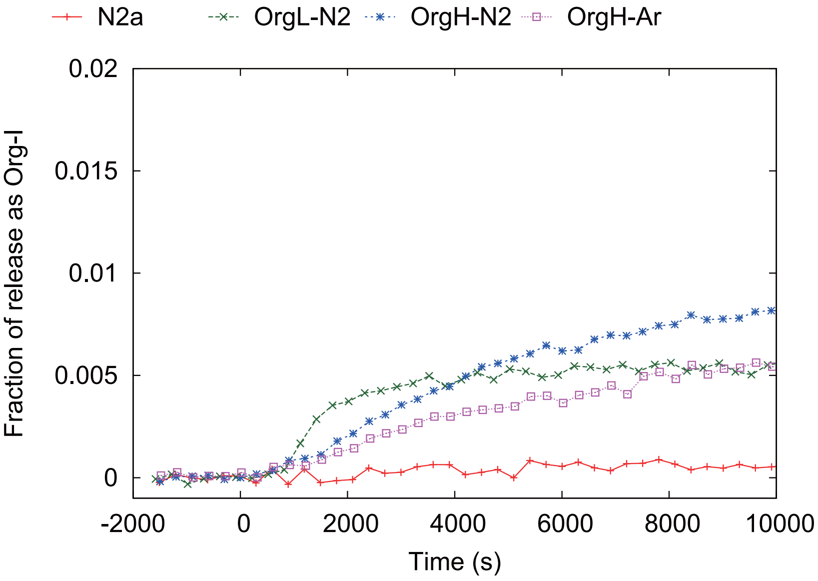


(b) Evolution of release along time.

Figure 2.16: Evolution of gaseous organic iodine release: influence of organic impurity (MIBK) and oxygen concentration. (air and low oxygen conditions)

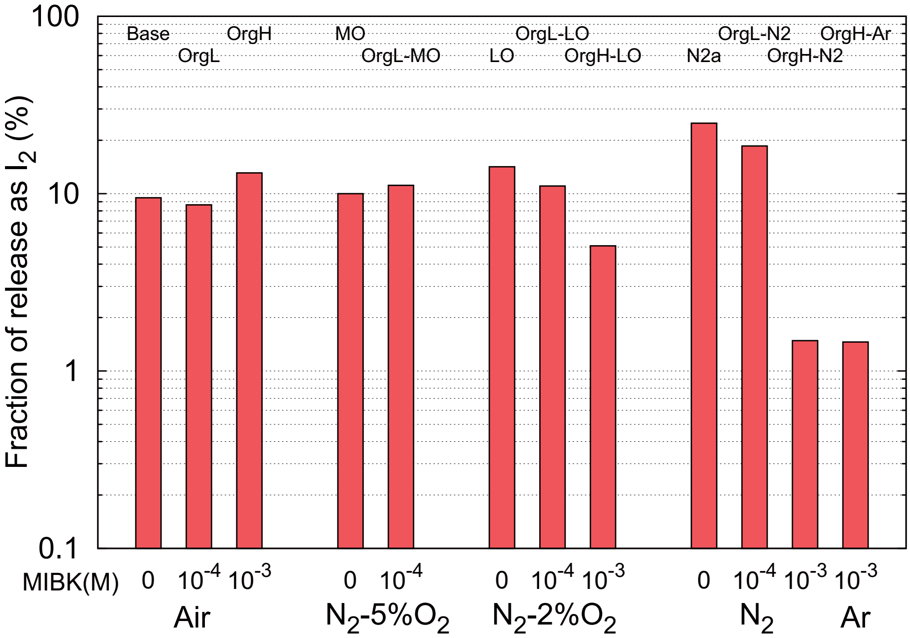


(a) Evolution of release in terms of absorbed dose.

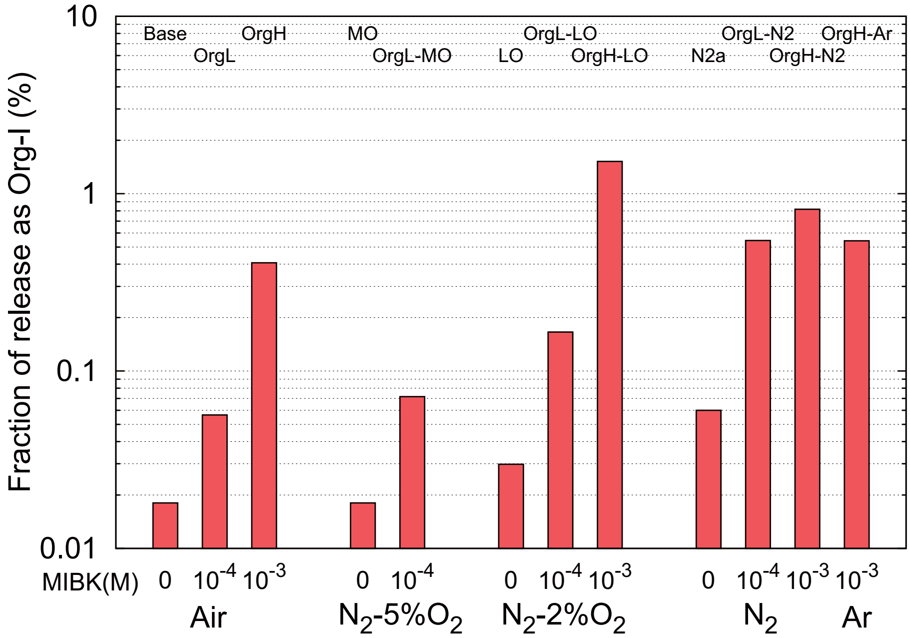


(b) Evolution of release along time.

Figure 2.17: Evolution of gaseous organic iodine release: influence of organic impurity (MIBK) and oxygen concentration. (no oxygen conditions)

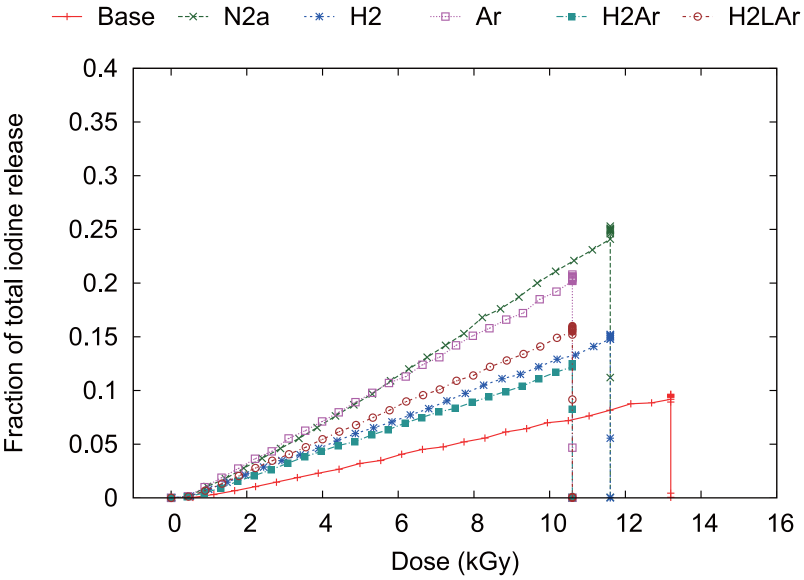


(a) Release as I_2.

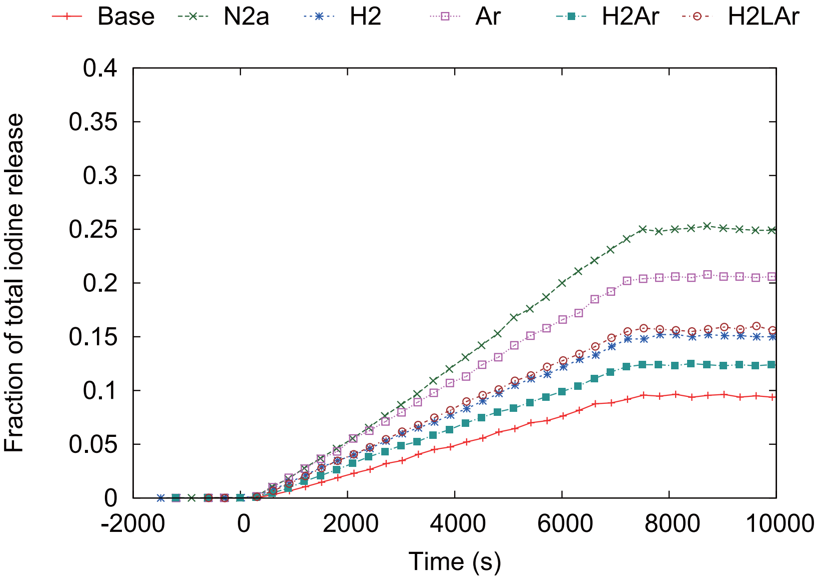


(b) Release as organic iodines.

Figure 2.18: Comparison of the final release fractions: influence of organic impurity (MIBK) and oxygen concentration.



(a) Evolution of release in terms of absorbed dose.



(b) Evolution of release along time.

Figure 2.19: Evolution of total gaseous iodine release: influence of hydrogen.

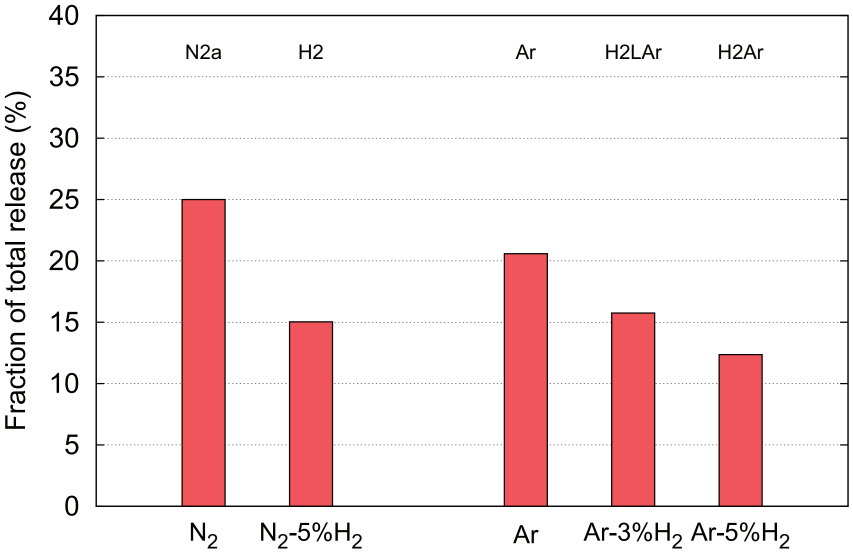


Figure 2.20: Comparison of the final release fractions (total iodine): influence of hydrogen.

3 Cold Experiments

3.1 Suppression of Iodine Release by Ammonia Alkaline

3.1.1 Purpose

As an accident management (AM) measure for the hydrogen accumulation issue in BWRs, Hamazaki et al.[22] proposed reduction or annihilation of hydrogen in the CVs by combining it with nitrogen through catalytic reactions producing ammonia. Ammonia is highly soluble in water, thus most of the hydrogen in the gas phase can be absorbed in water when converted to ammonia. Aqueous solution of ammonia is alkaline, and it is also expected that this method brings suppression of iodine volatility. However, considering that chemical reactions of ammonia or ammonium ions in the water pool may cause various influences, further study on the proposed AM measure including experimental work is necessary.

We performed experiments on the stability of ammonia alkaline solution containing iodine under irradiation.

3.1.2 Methods and Conditions

The possible maximum ammonia concentration in the suppression pool in BWRs by the proposed AM measure is obtained from the amount of hydrogen in the CV. The reaction of ammonia production,

$$N_2 + 3H_2 \longrightarrow 2NH_3 \ , \tag{3.1}$$

and the typical partial pressures of hydrogen and nitrogen, ∼0.1 MPa for both, lead to that it is likely to be excess nitrogen condition and ammonia production is 2/3 of the hydrogen inventory, ∼0.067 MPa. Based on the gas and water volumes in BWR CVs, the ammonia concentration in the suppression pool is about 0.1 M at maximum. We took this concentration for the standard case in our experiment. Other conditions were similar to that of the gaseous iodine release experiments described in the previous chapter.

Figure 3.1 illustrates the experimental system. Table 3.1 summarizes the conditions and results of the experiment. Note that this series was performed without radio active iodine, and all the measurement was by chemical means.

The test solution was 1×10^{-4} M CsI in ammonia alkaline conditions. Ammonia concentration was 0.1 M or 0.01 M. The sweep gas was nitrogen except one case swept with 5% oxygen-nitrogen mixture.

The test solution, 100 mL, was prepared in a 300 mL (nominal) separable flask. It was closed, loaded in the irradiator and connected to the sweep line. Then, sweep (flow rate 1 L/min), stir (6 mm(dia.) by 25 mm long glass stir bar, 350 rpm) and irradiation were given. The irradiation was given for 2 h at 5.1 Gy/h. The case 22-C02 was a reference case without irradiation.

In the cases 22-C02–C04, the pure water used as solvent was purged of oxygen with nitrogen for 30 min in advance, before adding ammonia and iodide to the prescribed concentration. The sweep was given for the period of irradiation.

In the case 22-C05, the test solution prepared with fresh pure water was loaded, and the sweep started 30 min before the start of irradiation so that the solution became saturated with the 5% oxygen gas at the start of irradiation. Also, the sweep continued extra 30 min after the end of irradiation. So, the sweep period was 1 h longer than other cases.

The iodine release from the solution was measured by bubbling the outlet gas in an alkaline (0.1 M NaOH) trap and monitoring iodide concentration with an ion selective electrode (ISE)

Table 3.1: Experimental conditions and summary of the results: ammonia effect experiments

No.	Preparation conc. (M)		Sweep gas	Solution pH (measured in test vessel)		I^- conc.($\times10^{-4}$M) (by sample)*1		NH_3 conc.(M) (by sample)*1		Fraction remained in solution*2		Fraction in trap		NH_3 loss fraction	Notes
	I^-	NH_3		Initial	Final	Initial	Final	Initial	Final	I^-	NH_3	I^- *3	NH_3		
22-C02	1.0E-4	0.1	N_2	11.10	10.81	1.14	1.25	0.0928	0.0450	1.07	0.475	3.5E-3	0.46	0.065	No irradiation
22-C03	1.0E-4	0.1	N_2	11.19	10.89	1.09	1.14	0.103	0.0574	1.02	0.544	2.8E-3	0.44	0.016	
22-C04	1.0E-4	0.01	N_2	10.64	10.28	1.03	1.06	0.00798	0.00396	1.00	0.483	8.9E-4	0.41	0.107	
22-C05	1.0E-4	0.1	N_2-5%O_2	11.24	10.70	1.28	1.24	0.106	0.0399	0.94	0.363	-2.4E-3	0.58	0.059	Extra 30 min gas purge before and after irradiation

*1) The measurement and the probe calibration were both done in 0.1M NaOH base solutions; samples were 10 times diluted with the base solution.
*2) Based on measurements by electrodes; decrement of the solution due to evaporation was considered.
*3) I^- concentrations were measured by compensating the sensitivity of the probes to ammonia.

Common conditions: Temperature=20-25°C, Vessel=300mL(nominal) separable flasks, Stir=ϕ6x25mm glass stirrer 350rpm, Volume of the test solution=0.1L, Gas flow rate=1L/min, Dose=5.1kGy/h x 2h, Trap=0.1M NaOH 0.25L (the gas was bubbled in the trap)
In C03 and C04, the the water was purged of dissolved air with N_2 for 30 minutes in advance; In C05, the vessel with the prepared solution was purged with the prescribed gas in extra 30 minutes before and after the irradiation (ammonia release was larger due to this)

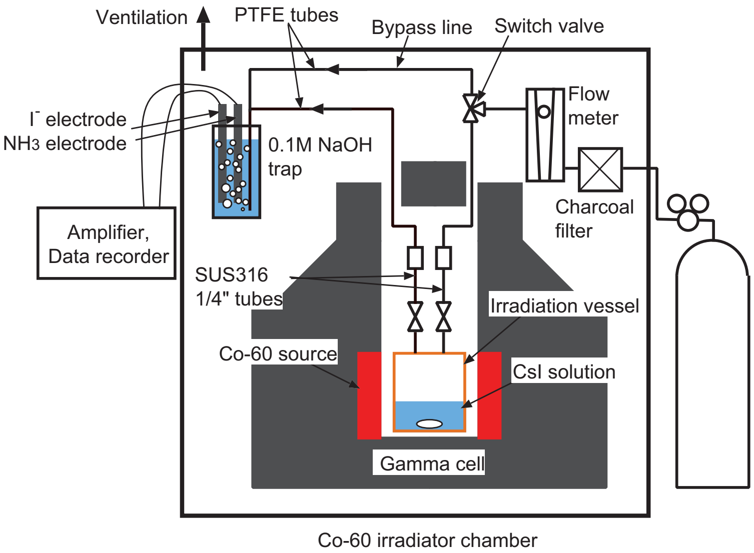


Figure 3.1: Schematic illustration of the experimental system.

(9653, Thermo) there. It is the same method as in the measurement of I_2 transfer in Appendix A, and the detail is described there. The release of ammonia from the solution was also monitored by inserting an ammonia electrode (9512, Thermo) to the trap. The ammonia electrode needs alkaline condition (pH>12) to detect the total of ammonia and ammonium and it was satisfied by 0.1 M NaOH in the trap.

A problem arose here was that the iodide electrode had a sensitivity on ammonia which was significant when the iodide concentration was low. As a workaround for this, we calibrated the iodide electrode for the influence of the existence of ammonia as in Fig. 3.2. The figure (a) shows the output voltage of the electrode with changing ammonia concentration at iodide concentrations 0 and 5×10^{-7} M. The result was fitted with a modified Nernst formula,

$$E = E_0 + A\frac{2.30\,RT}{nF}\log_{10}([\mathrm{I}^-] + f[\mathrm{NH}_3] + g) \tag{3.2}$$

where the variables denote

E, E_0 : output and standard voltages,

R : general ideal gas constant (= 8.31 J/mol K)

T : temperature (K)

n : charge of the ion concerned ($= -1$ for I^-),

F : Faraday's constant ($\equiv N_A\, e = 9.65 \times 10^4$ C/mol),

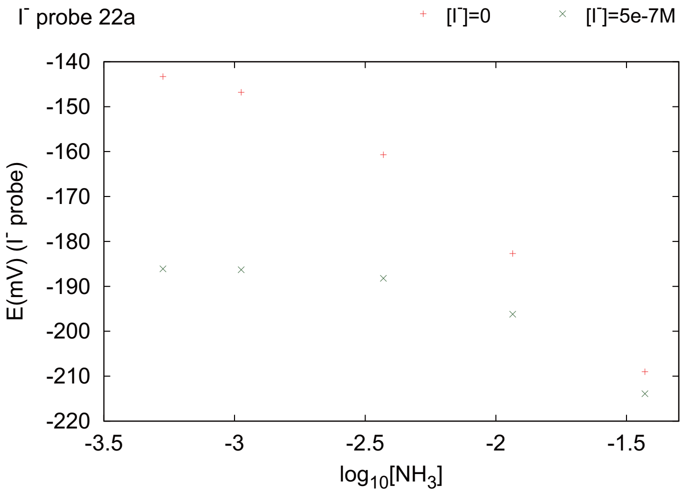
$[\mathrm{I}^-]$, $[\mathrm{NH}_3]$: concentrations of I^- and NH_3,

A : calibration constant without influence of ammonia,

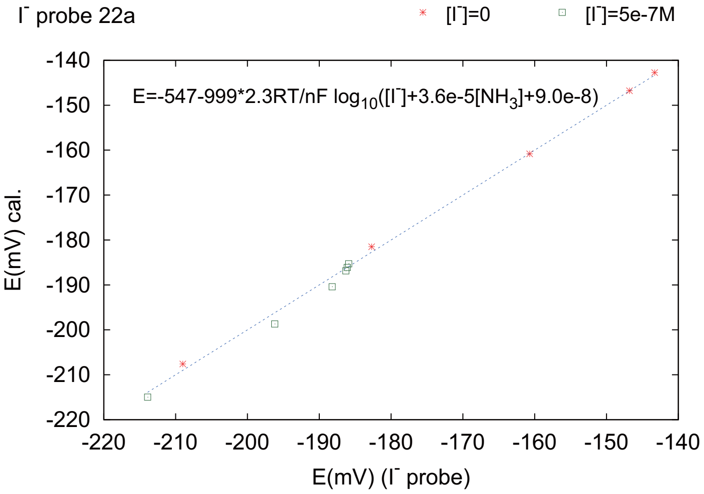
f, g : constants for the compensation for ammonia influence.

As shown in Fig. 3.2 (b), the ammonia influence was compensated well.

The pH, iodide and ammonia concentrations in the test solution at the initial and final states were also measured. The pH was measured by inserting an pH electrode (8102, Thermo) directly into the test solution before and after a run. Iodide and ammonia concentrations were measured by taking 2 mL samples before and after a run. The samples were diluted by ten times in 0.1 M



(a) Influence of ammonia on the iodide ion electrode output at low iodide concentrations.



(b) Calibration of the iodide electrode for the compensation of the ammonia influence.

Figure 3.2: Influence of ammonia on iodide ion electrode (9653, Thermo) and compensation.

NaOH solution, and measured by the electrodes. It was to make the condition of the samples consistent with that for calibration.

3.1.3 Results

The result for each case is given in Appendix B in detail. The summary of the result in Table 3.1 gives following observations.

- The pH by ammonia alkaline in the standard cases was initially 11, and dropped by 0.3–0.5 during the run by the transfer of ammonia. The initial pH was lower when ammonia concentration was lower (22-C04, 0.01 M). The pH drop was larger when sweep period was longer (22-C05, 1 h longer).
- The fractions of ammonia transferred was about 50%.
- The fractions of iodine released was in the order of 0.1% according to the trap iodide monitoring. It corresponded to concentrations $\sim 10^{-7}$ mol, in the order of the detection limit. The iodine concentration measurement for the test solution did not give significant change.
- No significant difference was observed between the irradiated and not irradiated cases (22-C02 and 22-C03).

Thus, it was concluded from this series of experiments that ammonia alkaline solution well kept iodide from releasing by radiolytic reactions and the irradiation did not affected the effect significantly.

More detailed examination, especially in a prototypic conditions, should be necessary if the ammonia production method for hydrogen/iodine control in BWRs is practically considered.

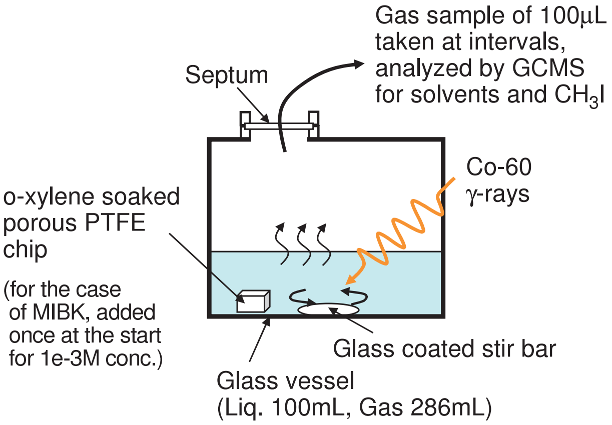


Figure 3.3: Schematic illustration of the xylene-MIBK comparison experiment.

3.2 Comparison of the Influence of Organic Compounds: MIBK and Xylene

3.2.1 Purpose

In the gaseous iodine release experiments with organic additives (the previous chapter), we used MIBK as one of organic compounds likely to leach from the paints into the water pool in CVs, partly because it had relatively high gas-liquid partition coefficient (low volatility) and was convenient for a good control on the organic concentration in the experiment.

On the other hand, some products of epoxy paints used in CVs are known to contain other types of organic solvents, mainly xylene[23, 24]. Therefore, we performed experiments to see if there was a significant difference between MIBK and xylene in terms of the influence on the gaseous iodine production by radiolysis.

3.2.2 Methods and Conditions

A difficulty in using xylene in the gaseous iodine release experiment is its low gas-aqueous partition coefficient ($H = C_l/C_g$ at equilibrium), $\sim$6 (o-xylene), compared with $\sim$400 of MIBK at 25 °C. Also, xylene has a very low aqueous solubility, 1.6×10^{-3} M at 25 °C, compared with 0.18 M for MIBK. In the experiments with MIBK, a high concentration solution, 0.1 M, was injected to the test vessel before the irradiation so that required concentration, (10^{-4}–10^{-3} M) was obtained in the test solution. The loss of MIBK by the transfer due to the sweep was not critical. However, we can not prepare a concentrated xylene solution nor keep xylene in the solution at sufficient concentration during irradiation with sweeping.

We adopted a method using a piece of porous PTFE material which absorbs xylene and releases it slowly when submerged in water. Figure 3.3 shows the method of the experiment. The experiment was done without sweeping the cover gas; the test vessel was closed. Gaseous samples were taken at intervals and analyzed for concentrations of xylene and CH_3I as a typical organic iodine. Among three isomers of xylene, we used o-xylene that has a larger gas-aqueous partition coefficient, $\sim$6 at 25 °C, compared to $\sim$4 for m- and p-xylene[25].

We did a similar experiment with MIBK as a reference case, with the same closed condition. We tried the method using the porous PTFE chip for MIBK, too. And, we found that the absorption and release of MIBK to/from the PTFE chip was too quick, almost fully absorbed or released within 1 h. So, we simply added MIBK once at the start of irradiation to give a desired concentration.

Table 3.2: Experimental conditions and summary of the results: comparison of xylene and MIBK

Name	No.	Organic addition	pH		Final I^- remaining fraction in solution* (%)	Gaseous CH_3I fraction*(%)
			Initial	Final*		
OrgCX	22-C10	o-Xylene (5E-4M at the irradiation start, leach from porous PTFE chip immersed in the solution 1h before the irradiation start)	6.91	5.78	70	0.18
OrgCM	22-C11	MIBK (1E-3M at the irradiation start)	6.92	6.2	81	0.25
Common condition	Solution: 1E-4M CsI, pH buffer: Boric acid-NaOH Irradiation: 5.0kGy/h × 2h Vessel: 300mL(nominal) separable flask, Liquid 100mL/Gas 286mL (closed) Cover gas: Air, Temperature: 25°C, Stir: ϕ6mm x 25mm glass stirrer 350rpm					
GCMS Analysis condition	Column: Agilent HP-5 (slightly polar, 320μm inner diameter, 0.25μm thick film, 30m long) Carrier gas: He, 1.3mL/min, Sample volume: 100μL, Split ratio: 1 Temperature control: 40 °C 4 min, 20 °C/min ramp up to 200 °C SIM (selected ion monitoring): m/z specification for target materials in three time rages of their release (CH3I:141,127,142, 40s--2m10s; MIBK:100,85,58,57,43, 2m11s--5m; o-Xylene:106,105,91,5m--11m10s) Calibration: CH3I: Standard (SPELCO 2000ppm) diluted solution confined in a vial, kept at a constant temperature, 25 °C; the gas phase sample was used; the known partition coefficient 4.82 (Glowa et al.,2003) was referred. o-Xylene: A small amount of the reagent (Wako, >98%) confined in a vial , kept at prescribed temperatures; the gas phase sample was used; the known vapor pressure (by Cox-Antoine formula) was refferred. MIBK: The initial concentration of the test solution (1E-3M) and known partition coefficient (Dewulf et al., 1995) were referred.					

* At the end of the test (samples were taken 30 min after the end of the irradiation)

Table 3.2 summarizes the experimental conditions and results for the two cases with xylene and MIBK.

The analysis of gaseous samples for the solvents, o-xylene or MIBK, and for CH_3I was done with gas chromatograph mass spectrometry (GCMS) (JMS-Q1000GC/K9, JEOL). The conditions of GCMS analysis are also included in the table. Gas phase samples of 100 μL were taken several times during a run by syringe through the septum in the lid of the vessel, and analyzed immediately. A simultaneous analysis of CH_3I and the solvents were not straight forward due to the large difference of their concentration ranges. It was enabled by a grouped selected ion monitoring (SIM) method in which the mass-charge ratio (m/z) for the SIM was set differently for specific zones of the retention time for different target molecules, namely, SIM for CH_3I was set for retention time 40s–2m10s, MIBK for 2m11s–5m, o-xylene after 5m. The calibration of GCMS for quantitative analysis was based on known gas phase concentrations. For CH_3I and o-xylene, the gas phase in vials containing standard solutions of CH_3I and pure o-xylene liquid, respectively, at given temperatures were used as standards. For the MIBK, the known initial gaseous concentration of MIBK at the start of the run from the given aqueous concentration and the gas-aqueous partition coefficient was referred. The gas-aqueous partition coefficients of these materials and the vapor pressure of o-xylene was obtained by the following equations. They were used also in processing the data.

- Gas-liquid partition coefficients
  - CH_3I[26] : $H = 4.82 \exp\left\{3420\left(\frac{1}{T} - \frac{1}{298}\right)\right\}$
    (T : temperature (K))
  - MIBK[27] : $H = RT \exp\left(-\frac{\Delta H^0 - T\Delta S^0}{RT}\right)$
    (R : general idea gas constant = 8.31 (J/mol K), T : temperature (K), $\Delta H^0 = -59$ (kJ/mol), ΔS^0 = -214 (J/mol K))
  - o-xylene[25] : $\ln \frac{1}{H} = \frac{a}{T} + c$
    (T : temperature (K), $a = -4232$, $c = 12.4$)
- Vapor pressure
  - o-xylene[28] (Cox-Antoine formula) : $\log_{10} P = A + B/(t + C)$
    (P : vapor pressure (mmHg), t : temperature (°C), $A = 9.28489$, $B = -3492.89$, $C = 401.824$)

We did not use a radioactive tracer in these experiments, and the gaseous partition of iodine was examined solely for CH_3I, a typical volatile organic iodine. The solution pH and iodide concentartion were measured by electrodes (8102 and 9653, Thermo) before and after a run.

The porous PTFE material (PTFE filter sheet 6 mm thick, Flon Industry) was cut into a 1×1 cm, 6 mm thick chip. The porosity of the material, evaluated by the comparison of its superficial density and the solid density of PTFE[29], was about 30%. Its absorption and release of o-xylene was tested in advance by a “pre-test”. The chip absorbed o-xylene to the amount filling out the pore in about 600 h when submerged in o-xylene. The release of o-xylene when the chip was dipped in water was measured under a similar condition to the experiment, *i.e.* 100 mL water in the test vessel at 25 °C, 350 rpm stir. The result showed that the o-xylene concentration in the solution was about 5×10^{-4} M at 1 h after the chip was put in the solution.

The following shows the procedure and schedule of the experiments. The time 0 is the start of irradiation.

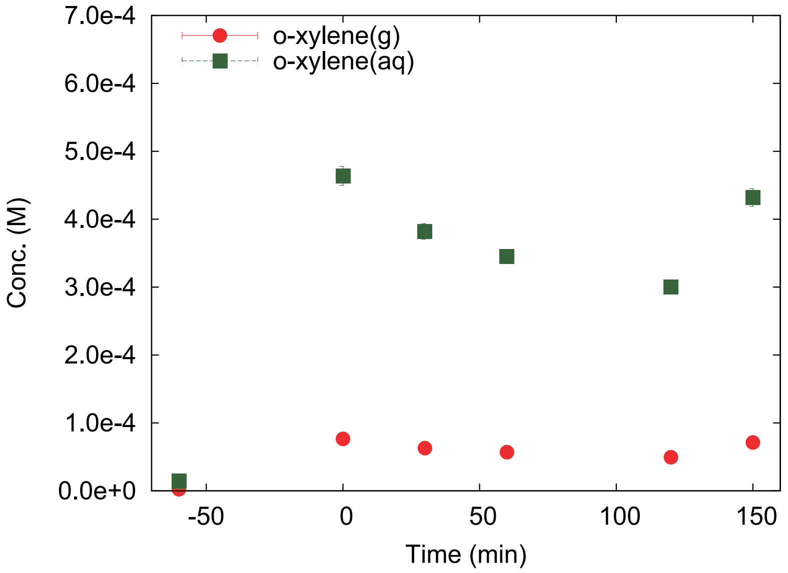


Figure 3.4: Evolution of the o-xylene concentration: OrgCX (irradiation 0–120 min).

−60 min	PTFE chip containing o-xylene was put in the solution, and a gas sample was taken (OrgCX)
0 min	MIBK was added to the solution to the conc. 1×10^{-3} M (OrgCM)
	Irradiation started
30 min	Irradiation interrupted; A gas sample was taken
60 min	Irradiation interrupted; A gas sample was taken
120 min	Irradiation ended; A gas sample was taken
150 min	A gas sample was taken

3.2.3 Results

Figure 3.4 shows the evolution of o-xylene concentration during the experiment OrgCX. The gas phase concentration is the measured value, and the aqueous phase concentration is the calculated value by assuming gas-aqueous equilibrium. The xylene concentration rose during the period before the start of irradiation, decreased during the irradiation due to radiolytic decomposition, and rose again after the end of irradiation.

An estimation of the released o-xylene amount was given by referring the o-xylene release behavior in the pre-test. Figure 3.5 compares the o-xylene release behavior in the pre-test and in the OrgCX case. Note that the data are the total released o-xylene mass (mol). The evolution in the first 60 min was similar with a slightly higher value in the OrgCX case. The curves are the one fitted to the pre-test data, and the other modified for fitting the initial 60 min of OrgCX data with the same time constant. Thus, the curve, $4.7 \times 10^{-4}\{1 - \exp(-2.6 \times 10^{-3}t)\}$ (t: time (min)) gives the o-xylene release history applicable for OrgCX, presumably in a computer simulation of the experiment.

Figre 3.6 shows the fraction of iodine partitioned in the gas phase as CH_3I in OrgCX. The error bars indicate the uncertainty evaluated based on scattering of the calibration data, $\sim 60\%$. The result showed a detectable production of CH_3I during the irradiation, in the range of 0.1% of

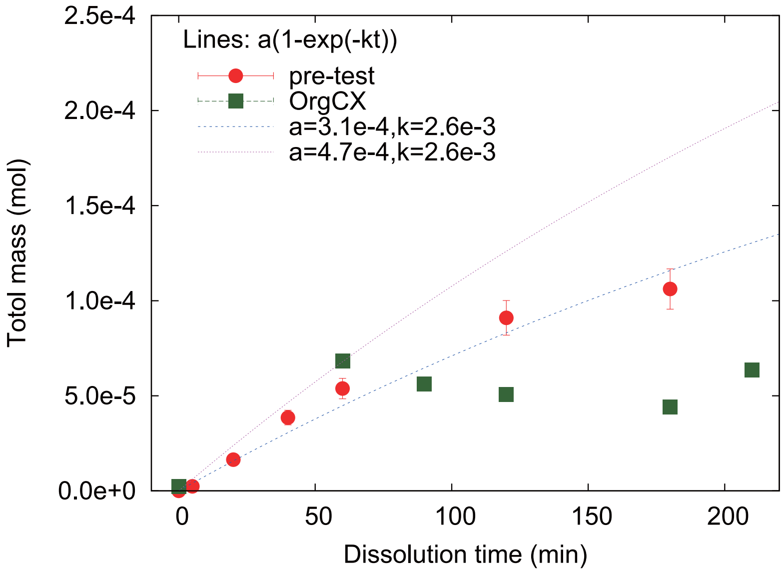


Figure 3.5: Comparison of the xylene mass (mol) evolution in the pre-test (no-irradiation) and the case OrgCX (irradiation 60–180 min: time from the immersion of the PTFE chip).

the initial iodide inventory. The aqueous phase values were deduced from the gas phase analysis results and the partition coefficient.

Figures 3.7 and 3.8 show the evolution of MIBK concentration and CH_3I in the case OrgCM. Most of MIBK was decomposed and detectable amount of CH_3I was produced. The fraction of iodine partitioned in the gas phase as CH_3I was comparable to that in the OrgCX case.

Comparing two cases, OrgCX and OrgCM, we do not see significant difference in the influence of o-xylene and MIBK in terms of organic iodine production.

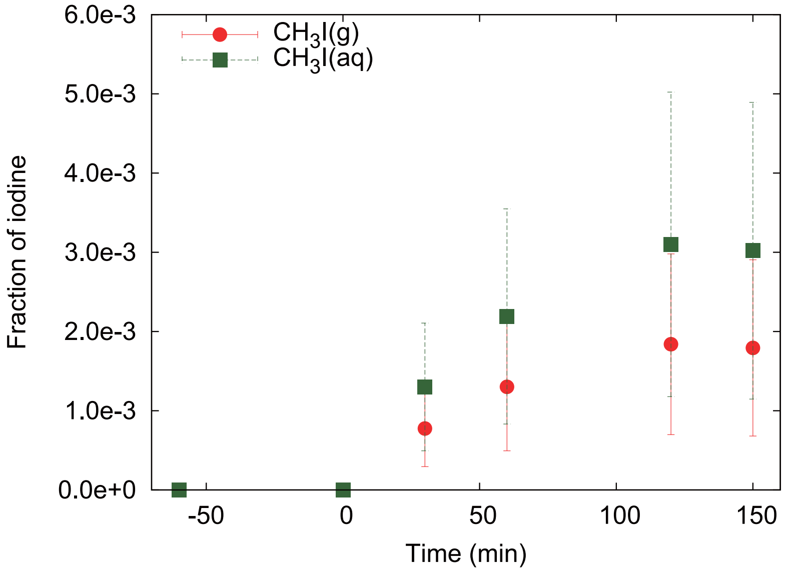


Figure 3.6: Evolution of the CH_3I concentration: OrgCX (irradiation 0–120 min).

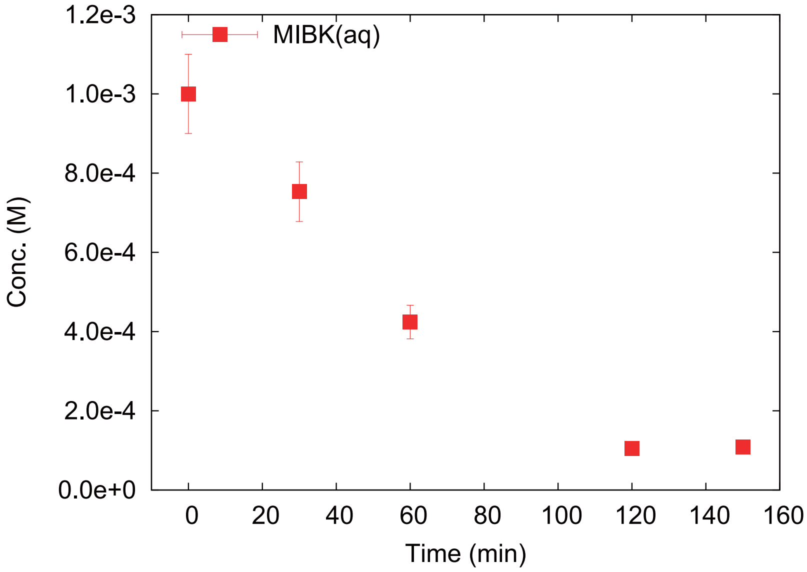


Figure 3.7: Evolution of the MIBK concentration: OrgCM (irradiation 0–120 min).

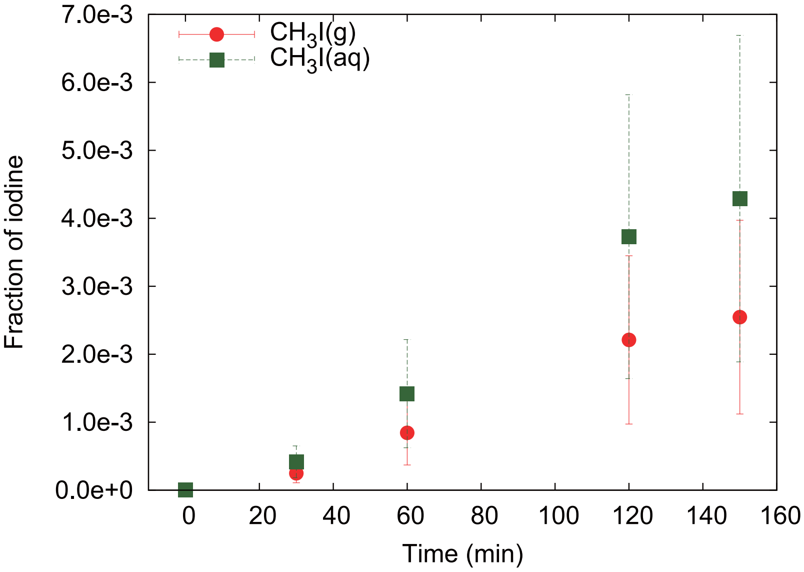


Figure 3.8: Evolution of the CH_3I concentration: OrgCM (irradiation 0–120 min).

4 Summary

The results of the present work are summarized as follows.

Gaseous iodine release experiment The volatile iodine release from γ-irradiated CsI solutions (concentration 10^{-4} M,^{131}I-labeled) was experimentally examined with a small scale and well controlled experimental apparatus, in which the test solution was kept at a constant temperature and stirred, the cover gas over the test solution was continuously swept, the released gaseous iodine was collected in the downstream and quantified. By collecting gaseous inorganic iodine (I_2) and organic iodines separately with species selective filters, their time dependent release behaviors were measured simultaneously.

Gaseous iodine release was observed during the irradiation. Parametric effects on the iodine release were examined for pH, temperature, inorganic impurities including iron and chlorine ions, a combined influence of organic impurity (MIBK) and oxygen concentration, and hydrogen concentration. The results showed the following:

- pH change 5.5–9 caused more than two orders of change in the iodine release, higher fractions with lower pH,
- temperature increase from 25 °C to 40 °C caused slight decrease of the iodine release, at both pH= 7 and 5.5,
- addition of ferrous ions to a concentration comparable to that of iodide did not show significant influence,
- addition of chlorine ions to concentrations 100 times of that of iodide did not show significant influence,
- decrease of oxygen concentration in the cover gas caused increase of the iodine release,
- addition of organic impurities (MIBK) caused decrease of iodine release as I_2 when the oxygen concentration was low (<2%), and increase of organic iodine release,
- 5% of hydrogen in the cover gas caused decrease of the iodine release to about half of that without hydrogen.

Cold experiments The following experiments were performed without a tracer, and the analysis of iodine and other species were solely by chemical measures.

The stability of ammonia alkaline solution, pH$\sim$11, containing iodine as CsI under γ-irradiation was examined. The result showed very small fraction of iodine release, in the range of 0.1%. No significant influence of irradiation was detected.

A comparison of the influences of organic compounds, MIBK and o-xylene, on the organic iodine production was performed in a closed system. During γ-irradiation, partition of iodine in the gaseous phase as CH_3I was detected in both cases, in comparable fractions in the range 0.1% of the initial iodide inventory.

The gas-aqueous interface mass transfer coefficient in the present experimental apparatus was measured by complementary experiments as given in Appendix A. The initial and boundary conditions for each series of experiments are described in corresponding sections. Thus, the information required for computer simulation of the experiments are sufficiently provided, and, the present data are ready for references for such purposes.

Acknowledgments

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Appendix A Evaluation of the Interface Mass Transfer Coefficient

A.1 Purpose

When we want to obtain information about the liquid phase chemical processes from the gaseous iodine release experiment with a swept system as described in Chapter 2, we need to know the gas-liquid interface mass transfer coefficient to separate the effect of the chemical processes in the liquid and that of the inter face mass transfer. We performed the complementary experiment described in this section to suffice that need.

A.2 Method

The following equations express the definitions and relationships of variables concerned in the interface mass transfer based on the double boundary theory,

$$\begin{aligned} N &= k_g(C_g - C_{gi}) = -k_l(C_l - C_{li}) \;=\; K_{mt}(HC_g - C_l), && \text{(A.1)} \\ 1/K_{mt} &= H/k_g + 1/k_l. && \text{(A.2)} \end{aligned}$$

where, the variables denote

N : interface mass flux (mol/dm^{2}s),

C_g, C_l : bulk concentrations in the gas and the liquid phases (mol/dm^3),

C_{gi}, C_{li}: concentrations at the boundary in the gas side and the liquid side (mol/dm^3),

k_g, k_l : mass transfer coefficients at the gas side and the liquid side (dm/s),

K_{mt} : overall mass transfer coefficient (dm/s),

H : gas-liquid partition coefficient ($\equiv C_l/C_g$ at the equilibrium).

The inverse mass transfer coefficients, H/k_g, $1/k_l$ and $1/K_{mt}$, mean interface resistances in the gas side, in the liquid side and the overall, respectively. When the cover gas is swept like the present case, the gas phase is well mixed, and the mass transfer resistance ($1/K_{mt}$) is dominated by that in the liquid side, *e.g.* $1/K_{mt} \sim 1/k_l$. The liquid side mass transfer coefficient depends on the diffusion of the solute in the liquid as follows[30, 31],

$$k_l \sim D/\delta \quad \text{or} \quad k_l \sim D^n \quad (n = 0.5 \sim 1), \tag{A.3}$$

where D and δ denote the diffusion coefficient of the solute and the boundary layer thickness in the liquid side. The dependence of the mass transfer coefficient on the diffusivity, Eq.(A.3), is of the power $n = 0.5 \sim 1$. The diffusivity can be evaluated by the Wilke-Chang empirical equation[32, 33] for diffusivity of a dilute solute A in a solvent B,

$$D = 7.4 \times 10^{-8} \frac{(\psi_B M_B)^{0.5}\theta}{\mu V_A^{0.6}} \quad (\text{cm}^2/\text{s}), \tag{A.4}$$

where the variables denote

ψ_B : association parameter (= 2.6 for water),

M_B: molecular weight of the solute,

θ : temperature (K),

μ : viscosity of the solvent (approximately of water) (cp),

V_A : molecular volume of the solute as liquid at the normal boiling point (cm^3/mol).

This equation, reportedly giving the diffusivity of non-dissociating solutes within 10%[33], shows a dependence on temperature by $\sim \theta/\mu$ (assuming the solvent viscosity is a function of temperature) and on the solute material by $\sim V_A^{-0.6}$. Thus, the dependence of the overall mass transfer coefficient on these factors would be

$$K_{mt} \sim \frac{\theta}{\mu V_A^{0.6}} \tag{A.5}$$

by taking $n \sim 1$ in Eq.(A.3).

Assume a closed volume with gas and liquid phases, with an inlet and an exit in the gas phase through which the gas phase is swept by a constant flow. The evolution of the gas phase and liquid phase concentrations due to the interface mass transfer is expressed by the following,

$$\frac{dC_g}{dt} = \frac{A_i K_{mt}}{V_g}(C_l - HC_g) + \frac{q_g}{V_g}(C_{gin} - C_g), \tag{A.6}$$

$$\frac{dC_l}{dt} = -\frac{A_i K_{mt}}{V_l}(C_l - HC_g), \tag{A.7}$$

where A_i, V_g and V_l are the interface area, gas volume and the liquid volume, respectively; q_g and C_{gin} are the sweep gas flow rate (dm^3/s) and the concentration of the solute in the sweep gas at the inlet, respectively. When the concentration of the solute in the sweep gas is kept constant, $C_g \simeq C_{gin}$ =const., an approximated solution of the liquid phase concentration becomes

$$X = \exp(-K_{mt}T). \tag{A.8}$$

The variables X and T are normalized concentration and time, respectively, defined by

$$X = (C_l - HC_g)/(C_l^i - HC_g), \tag{A.9}$$

$$T = t(A_i/V_l), \tag{A.10}$$

where C_l^i is the initial liquid phase concentration. Thus, we can obtain K_{mt} by fitting the evolution of the liquid phase concentration to Eq.(A.8).

Figures A.1 and A.2 are the experimental set-up for the measurement of the evolution of liquid phase concentrations of oxygen and iodine, respectively. We measured the mass transfer coefficient of oxygen for many runs changing various parameters, and that of iodine for a standard condition.

In those experiments, test solution 100 mL was prepared in a 300 mL (nominal) separable flask and the flask was covered by the lid designed for the gaseous iodine experiment. The cover gas was swept by air or nitrogen and the solution was stirred by a glass coated 6 mm (dia.)× 25 mm stir bar during a run. We took as the standard condition the temperature 25°C, the stir revolution rate 350rpm and the gas flow rate 1L/min.

For the oxygen measurement, water was used as the test solution and a polarographic dissolved oxygen (DO) electrode (850A, Thermo) was inserted directly into the test water. For the iodine measurement, saturated solution of I_2 (about 10^{-3} M) was used as the test solution. The outlet gas from the vessel was bubbled into a trap containing NaOH 0.2 M solution. The

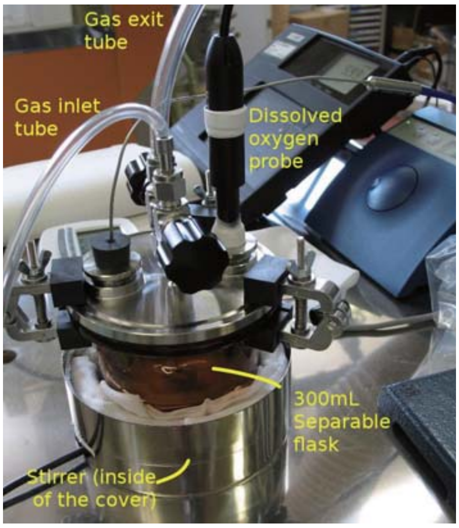


Figure A.1: Measurement of the interface mass transfer coefficient with oxygen.

gaseous I_2 transferred into the trap undergoes the following chemical equilibria.

$$\begin{aligned} I_2 + OH^- &\longleftrightarrow I_2OH^- \\ I_2OH^- &\longleftrightarrow HOI + I^- \\ HOI &\longleftrightarrow IO^- + H^+ \\ I_2 + I^- &\longleftrightarrow I_3^- \end{aligned}$$

At the pH of the trap solution (~13), practically, I_2 produces the same amount of I^-. Then, monitoring I^- concentration in the trap is equivalent to monitoring I_2 coming out of the test solution. An ion selective electrode (ISE) for iodide (9653, Thermo) was inserted into the trap to monitor the I^- concentration there.

The oxygen mass transfer was measured by the following procedures. At the start of every run, the vessel was swept by air until the test water became saturated with air. Next, the vessel was purged of oxygen with nitrogen, during which the oxygen concentration decrease was monitored. (transfer of oxygen from the water to the cover gas) Then, the vessel was swept with air again, and the increase of oxygen concentration was monitored. (transfer of oxygen from the cover gas to the water)

The iodine mass transfer was measured by sweeping the vessel containing the I_2 solution with air. The concentration of I^- in the trap was monitored during the run.

A.3 Results

Figure A.3 shows the measurement result of I^- concentration in the trap. The normalized I_2 concentration (Eq.(A.9)) in the test solution deduced from it is plotted against the normalized time (Eq.(A.10)) in Fig. A.4. The experimental result was well fitted by Eq.(A.9), and the overall mass transfer coefficient, $K_{mt} = 1.2 \times 10^{-4}$ dm/s, was obtained. Measurements on various samples were done for checking the mass balance of iodine as summarized in Table A.1. The trap efficiency obtained from the mass balance was considered in the data in Fig. A.4.

Table A.2 summarizes the results of the experiments. In the oxygen experiments, influences of various water conditions and procedures were examined because we suspected that small amount of impurities or other factors may affect the interface transfer. The result showed that

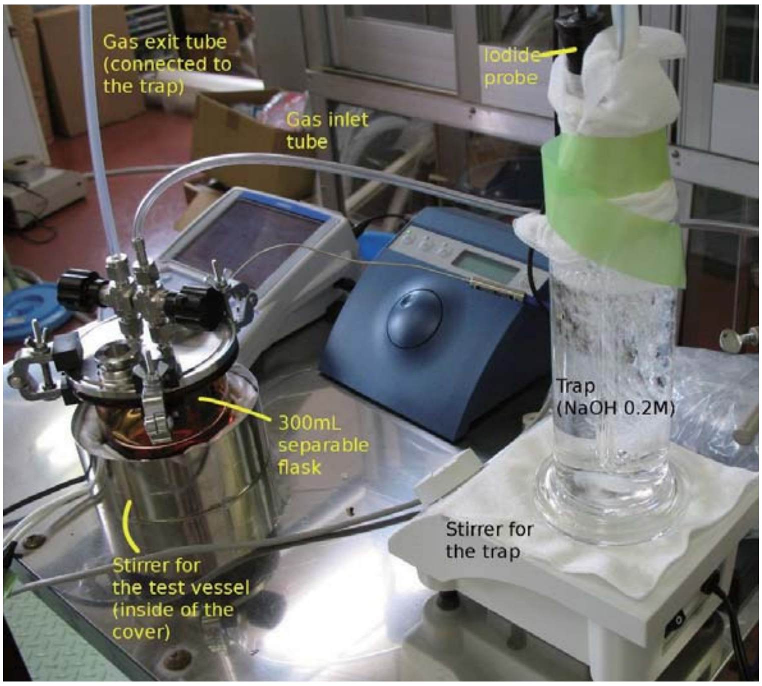


Figure A.2: Measurement of the interface mass transfer coefficient with iodine.

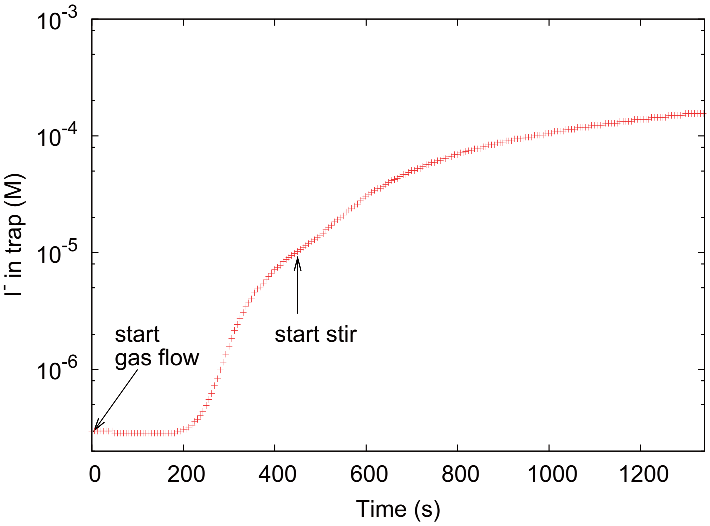


Figure A.3: Evolution of the iodide ion concentration in the trap.

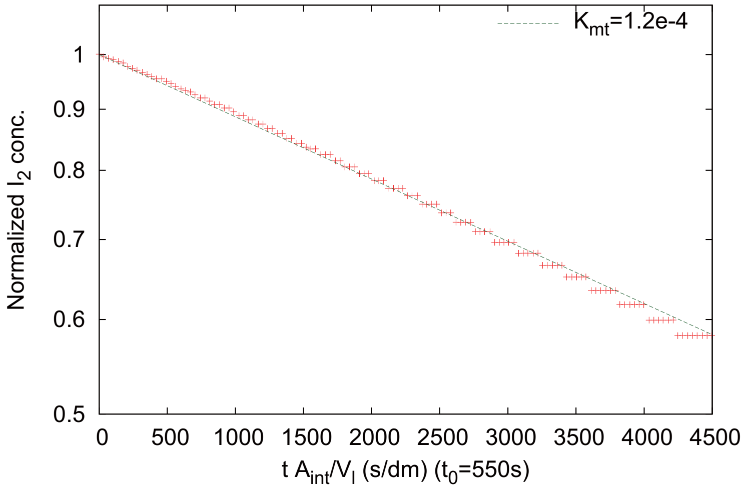


Figure A.4: Evolution of the normalized iodine concentration in the test solution.

Table A.1: Mass balance in the iodine mass transfer measurement.

Sample	Method of measurement	Conc.(M)/ Mass (mol)	Note
Test solution (before)	**The initial I_2 conc. based on the preparation**	**8.55E-04 (M)**	=A
Test solution (after)	I- conc. in the solution after I_2 release	1.57E-05 (M)	iodine remained in the solution as I-
	I- conc. in the solution after 5M NaOH 4mL was added and it turned basic (pH>13)	4.65E-04 (M)	
	The above conc. compensated for the volume increase	4.83E-04 (M)	I- after all I_2 was converted to I- in the basic condition
	Final I_2 conc. evaluated based on the equilibrium	**4.68E-04 (M)**	=B
Trap solution (0.2M NaOH)	I- conc. at the end	1.56E-04 (M)	
	I- mass in the trap (220mL)	3.42E-05 (mol)	
	The absorbed I_2 mass evaluated based on the equilibrium	**3.42E-05 (mol)**	=C
	Absorption efficiency of the trap, t_{reff}	**8.83E-01 (-)**	=C/{(A-B)x0.1}

the overall transfer coefficient, K_{mt}, mostly scattered in the range 2×10^{-4}—6×10^{-4} dm/s. The following trends were observed.

- Stirring by a magnet stirrer and its preriod did not affect.
- Fresh pure water (by Milli-Q, >18MΩm, TOC <30ppb) showed high transfer coefficients.
- CsI and methylethylketone (MEK) did not affect.
- Storing in a glass bottle did not affect.
- Exposure to the room air or storing in a plastic bottle may decrease the transfer coefficient.

Runs with pure water stored for a while in a glass bottle at the standard condition showed transfer coefficients around 4×10^{-4} dm/s with a good reproducibility. Then, influences of the temperature, stir revolution rate and gas flow rate were examined. Also, the difference between oxygen and iodine was examined in terms of the dependence indicated by Eq.(A.5). Among those factors, the influence of the gas flow rate was insignificant. The influence of other factors are shown in Figs. A.5–A.7. The trend was as follows. The stir revolution had nearly linear impact on the transfer coefficient. The dependence on the temperature, including the indirect effect through viscosity, was weaker than expected by Eq.(A.5) though they agreed qualitatively. The dependence on the molecular volume of the solute was stronger than expected by Eq.(A.5) though they agreed qualitatively. Considering those results, we added a linear dependence on the stir revolution $\sim R$, and developed the following empirical expression for the overall interface mass transfer coefficient in the present experimental system.

$$K_{mt} = K_{mt}^0 \frac{R}{R^0} \frac{(\theta/\mu)}{(\theta^0/\mu^0)} \left(\frac{V_A^0}{V_A} \right)^{0.6} \tag{A.11}$$

where the superscript 0 means the standard condition: oxygen, 25 °C and 350 rpm stir. The standard mass transfer coefficient is $K_{mt} = 4 \times 10^{-4}$ dm/s. The water viscosity μ (Pa·s) is given by

$$\mu = 1/(-2.759 \times 10^{-4}\,\theta^3 + 0.3562\theta^2 - 113.3\,\theta + 10550) \tag{A.12}$$

where θ is temperature (K). This expression is based on the data in Ref. [34], and valid for temperatures 274–450K.

Figure A.8 shows the comparison between the experimental data and the empirical equation Eq.(A.11). Table A.3 shows the molecular volumes of several materials for reference.

Table A.2: Measurement results of the interface mass transfer coefficient.

Run No.	Stir revolution (rpm)	Gas flow rate (SLM)	Temperature (℃)	K_{mt} (10^{-4} dm/s)	Note on the water quality etc.
1-8	350	1	23-27	1.8−2.0	water stored in a polyethylene bottle for 1 month
9-12,15-18	350	1	21-23	3.2−4.0	fresh pure water (18.2MΩm,TOC=14ppb)
13-14	350	1	23	3.9−4.2	1e-4M CsI
19-20	350	1	23	4.1−5.0	pure water stored in a glass beaker for 1 day
21-22	350	1	24-25	2.5−3.2	pure water stored in a polyethylene bottle for 1 day
25-26	350	1	21-22	4.1−5.1	fresh pure water
27-28	350	1	22	3.8−4.6	1e-5M MEK
29-30	350	1	23	3.6−4.2	1e-4M MEK
31-34	350	1	22	3.6−4.8	1e-3M MEK
35-44	350	1	21-23	4.4−5.6	no difference between fresh pure water and that exposed to the room air for 1.5h
A1-A2	350	1	20-22	3.7−3.9	pure water stored in a separable flask sealed with parafilm for 1 day
B1-B2	350	1	21	4.0−4.3	pure water stored in a separable flask sealed with parafilm for 1 day during stirred
C1-C2	350	1	20-21	3.5−3.8	pure water stored in a separable flask exposed to the room air for 1 day
D1-D2	350	1	21	4.0−4.4	water used in the case 44 was kept in a separable flask sealed for 1 day
E1,E2,Ea1,Ea2	350	1	22	5.2−6.3	fresh pure water
A@5/21a,b	350	1	18-19	4.6−5.6	pure water sealed in a glass bottle and kept in a dark place for 5 days
B@5/21a,b	350	1	21	4.7−5.8	fresh pure water
A@5/24a,b,a-re,b-re	350	1	22-24	3.7−4.4	pure water sealed in a glass bottle and kept in a dark place for 5 days and in a bright place for 3 days
A@5/24a420,b420	**420**	1	24	5.1−5.5	water used in the above
A@5/24a500,b500,a500-re,b500-re	**500**	1	23-24	6.1−6.6	water used in the above
A@5/24a2L,b2L	350	**2**	23-24	3.6−3.7	water used in the above
A@5/24a0.5L,b0.5L	350	**0.5**	24	3.6−3.7	water used in the above
A5/24a10C,b10C	350	1	**9***	2.0−2.4	water used in the above
A5/24a40C,b40C	350	1	**38**	4.4−5.1	water used in the above
Iodine transfer test (2007)	350	1	21	1.2	
Iodine transfer test (2006)	340	1	25	1.5−2	stir bar 11(dia.)-33mm
Iodine transfer test by Taghipour	110	4.2	RT	1.5	vessel 850mL
	380	4.2	RT	4.5	vessel 850mL

SLM: mass flow rate indicated by the volume flow rate at 0°C, 1 atm (L/min)
RT: room temperature=(20-25°C)
* : In the low temperature cases, the temperature in the vessel air was about 5°C higher than in the water; it might have caused some problem in the temperature compensation in the probe

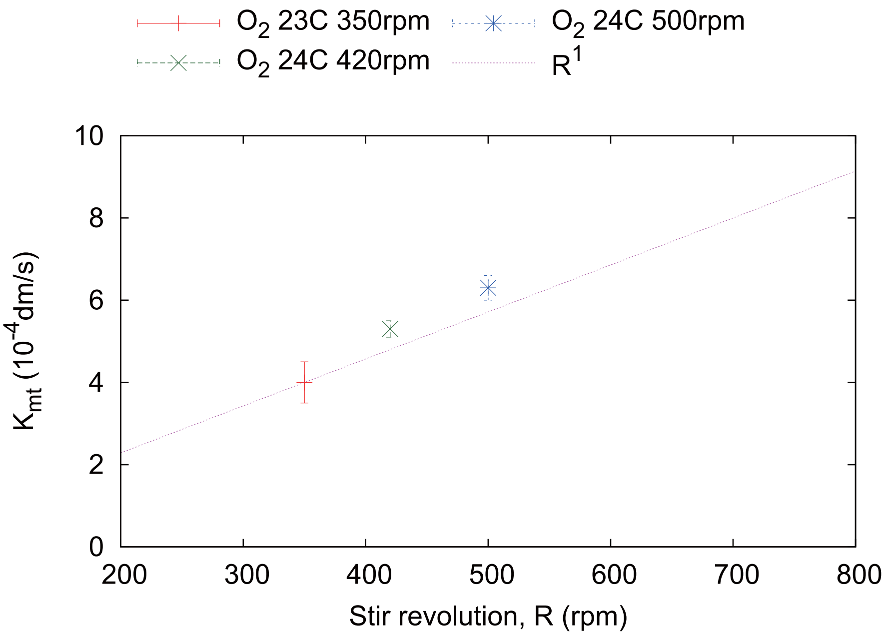


Figure A.5: Dependence of K_{mt} on the revolution of the stirrer.

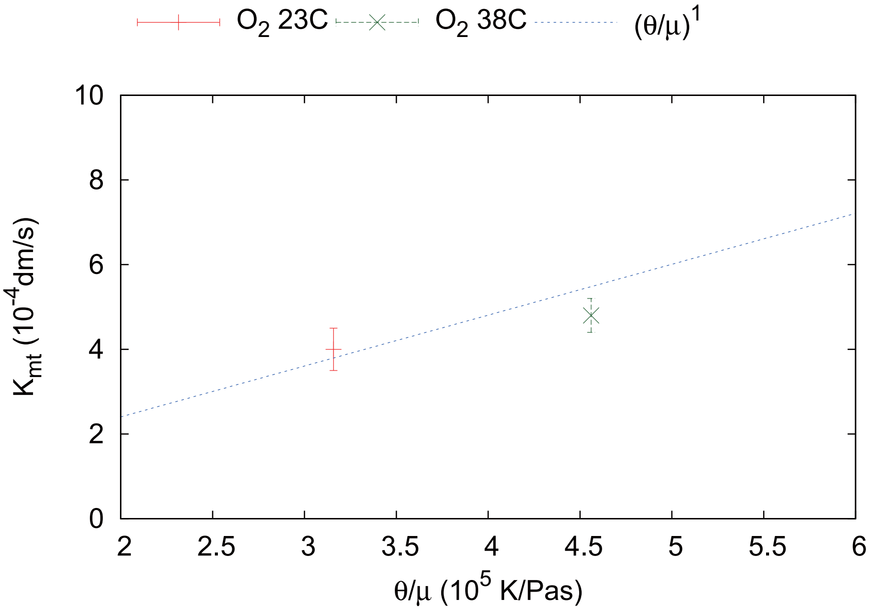


Figure A.6: Dependence of K_{mt} on temperature.

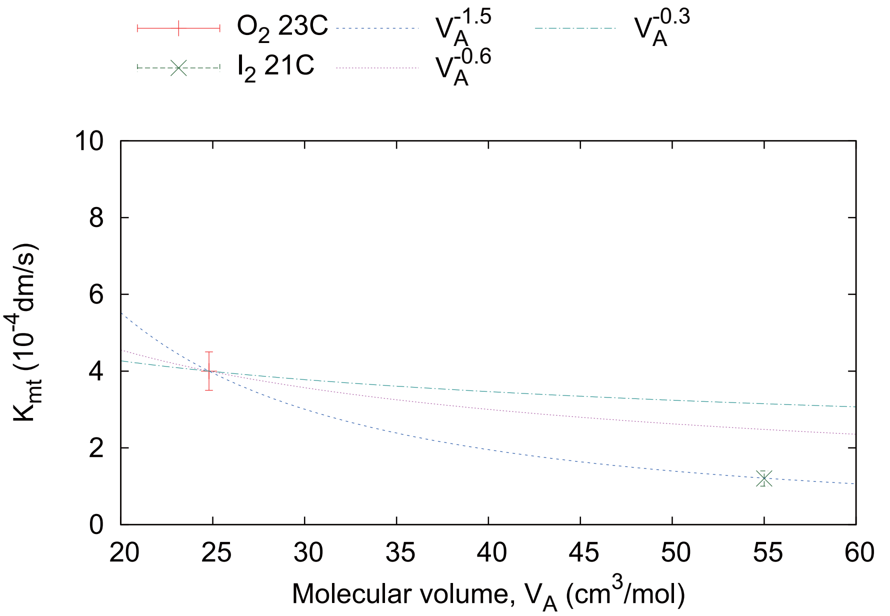


Figure A.7: Dependence of K_{mt} on the molar volume of volatile species.

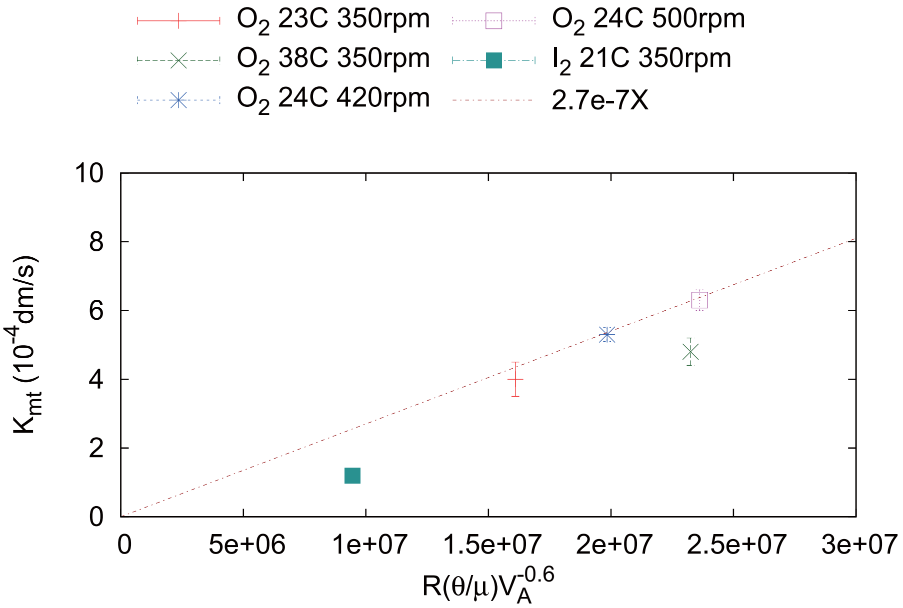


Figure A.8: Synthesized correlation for the dependence of K_{mt}.

Table A.3: Molecular volumes of various materials.[35, 36]

Material	Molecular weight	Density (g/cm^3)	Molecular volume (V_A) (cm^3/mol)	$1/V_A^{0.6}$	$1/V_A^{0.6}$ ratio to O_2	Note (condition for the referred density)
O_2	32.0	1.144	28.0	0.1355	1.000	−183°C
N_2	28.0	0.808	34.7	0.1192	0.879	−195.8°C
H_2	2.0	0.0708	28.2	0.1347	0.994	−253°C
I_2	253.8	3.710	68.4	0.0792	0.585	185°C
CH_3I	141.9	2.293	61.9	0.0841	0.621	18°C
C_2H_5I	156.0	1.929	80.9	0.0717	0.529	20°C
$CH_3(CH_2)_2I$	170.0	1.746	97.4	0.0641	0.473	20°C
$(CH_3)_2CHI$	170.0	1.704	99.8	0.0632	0.466	20°C

Appendix B Data Sheets

B.1 Gaseous Iodine Release Experiments

Experimental condition

Run No.	**19-01**	
Solution	pH=5.5 (Buffer: 0.1M B(OH)3-1.5e-5M NaOH), CsI 1e-4M, I-131 37kBq	
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar	
Gas	Air 1L/min	
Dose rate	7.60 kGy/h	
Date	2007/06/28	
Irradiation schedule		
	Sweep start	12:14
	Irrad. start	12:34
	Irrad. end	14:34
	Sweep end	15:04

Activity measurement

Run No.: 19-01 Date: 2007/06/28 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	07/06/28 12:51	300	31940	0.93	371.860	372.999	1.0000
Solution after irrad. 1mL	07/06/28 16:02	300	9802	1.86	114.127	115.796	3.10E-01
1: Ag-1	07/06/28 16:44	300	592960	0.23	18445.458	18762.523	5.03E-01
2: Ag-2	07/06/28 16:52	300	82682	0.62	2571.852	2617.317	7.02E-02
3: Ag-3	07/06/29 15:36	600	7164	2.61	111.423	123.065	3.30E-03
4: Ag-4	07/06/28 17:06	300	101	-	-	-	-
5: Ag-5	07/06/29 15:12	300	79	-	-	-	-
6: AgNO3-1	07/06/29 09:30	3600	5757	6.17	15.823	17.097	4.58E-04
7: AgNO3-2	07/06/29 14:54	300	29	-	-	-	-
8: AgNO3-3	07/06/29 15:02	300	164	-	-	-	-

* Fraction to total inventory

Activity values for solution samples were for whole solution

Sum for filter 1-8	5.769E-01
Frac. in solution	3.104E-01
Loss	1.126E-01

Chemical measurements

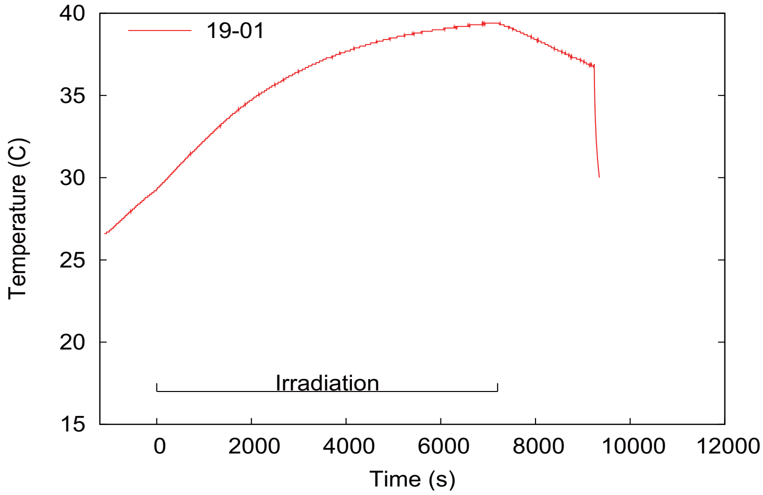
Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
19-01	5.355	5.629	9.98E-05	2.62E-05	2.62E-01

Filter fraction data

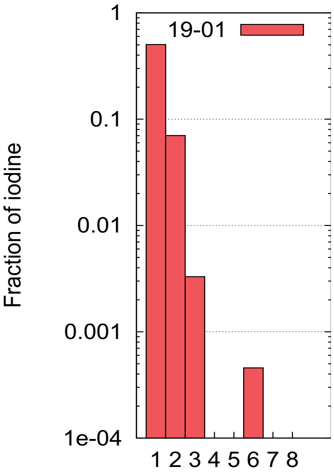
Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	0.503018	8.719E-01
2	0.070169	1.216E-01
3	0.003299	5.719E-03
4	-	-
5	-	-
AgNO3 (for Org-I)		
6	0.000458	7.945E-04
7	-	-
8	-	-

Plots

Temperature	Initial	26 ℃
	During irrad.	28--39 ℃

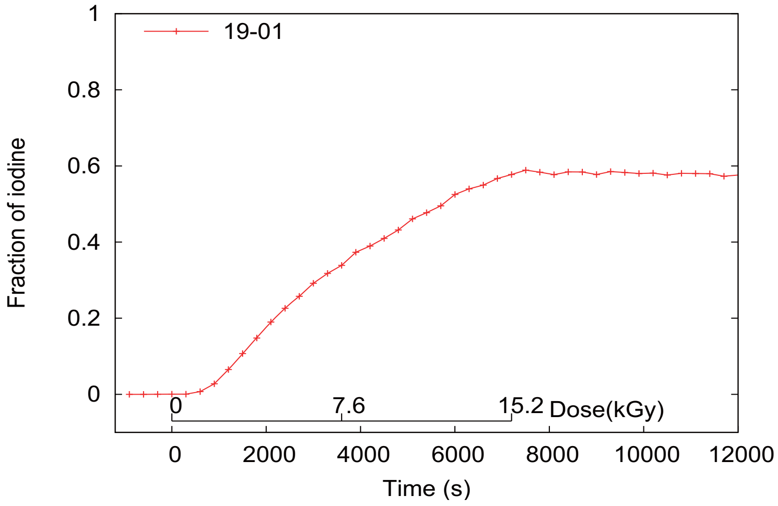


Distribution in filters



1-5: Ag filters
6-8: AgNO3 filters

Release history



Count efficiency(calibrated)=	0.1187

Experimental condition

Run No.	**19-02**
Solution	pH=7.0 (Buffer: 0.1M B(OH)3-5.72e-4M NaOH), CsI 1e-4M, I-131 37kBq
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar
Gas	Air 1L/min
Dose rate	7.44 kGy/h
Date	2007/08/29

Irradiation schedule

Sweep start	10:48
Irrad. start	11:08
Irrad. end	13:08
Sweep end	13:38

Activity measurement

Run No.: 19-02 Date: 2007/08/29 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	07/08/29 10:54	300	39780	0.79	463.136	461.305	1.000E+00
Solution after irrad. 1mL	07/08/29 14:32	300	36331	0.81	423.010	426.886	8.79E-01
1: Ag-1	07/08/29 15:36	300	89100	0.54	2771.671	2807.833	6.09E-02
2: Ag-2	07/08/29 15:42	300	43306	0.80	1347.138	1365.205	2.96E-02
3: Ag-3	07/08/29 15:54	600	8911	2.20	138.595	140.554	3.05E-03
4: Ag-4	07/08/29 16:33	600	1129	11.07	17.560	17.850	3.87E-04
5: Ag-5	07/08/29 16:45	600	702	16.95	10.918	11.107	2.41E-04
6: AgNO3-1	07/08/29 16:01	1800	5749	4.23	31.598	32.058	6.95E-04
7: AgNO3-2	07/08/29 16:56	300	-	-	-	-	-
8: AgNO3-3	07/08/29 17:01	300	-	-	-	-	-

Sum for filter 1-8	9.48E-02
Frac. in solution	8.79E-01
Loss	2.61E-02
Water loss by evaporation(%)	5.00

* Fraction to total inventory

Activity values for solution samples were for whole solution

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
19-02	6.746	6.673	9.98E-05	7.97E-05	7.98E-01

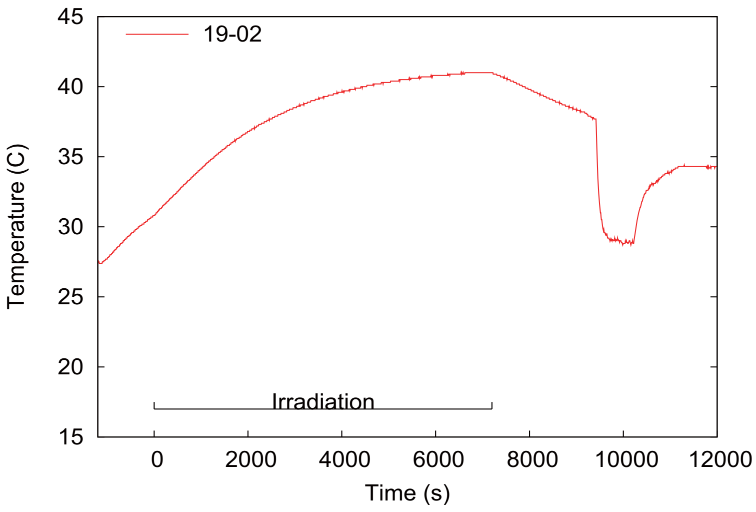
Filter fraction data

Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	6.087E-02	6.418E-01
2	2.959E-02	3.121E-01
3	3.047E-03	3.213E-02
4	3.869E-04	4.080E-03
5	2.408E-04	2.539E-03
AgNO3 (for Org-I)		
6	6.949E-04	7.328E-03
7	-	-
8	-	-

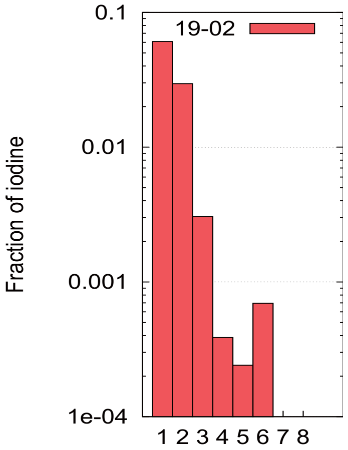
Plots

Temperature Initial 27 ℃

During irrad. 30--41 ℃



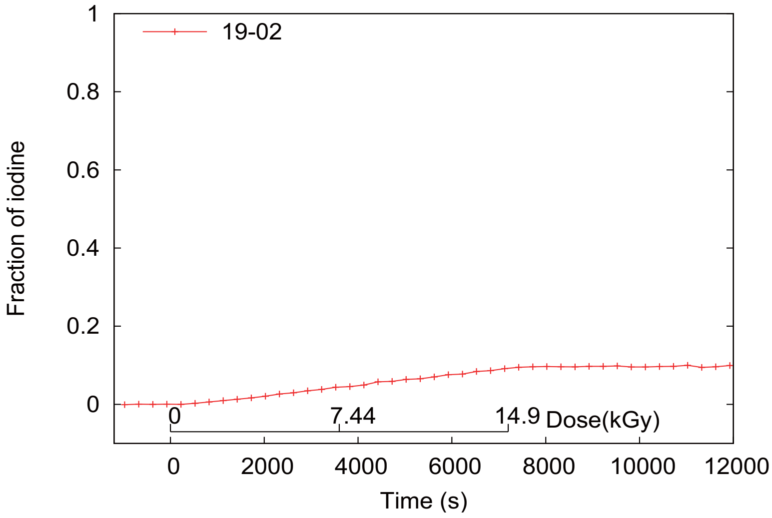
Distribution in filters



1-5: Ag filters

6-8: AgNO3 filters

Release history



Count efficiency(calibrated)= 0.1146

Experimental condition

Run No.	**19-03**		
Solution	pH=9.0 (Buffer: 0.1M B(OH)3-3.65e-2M NaOH), CsI 1e-4M, I-131 37kBq		
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar		
Gas	Air 1L/min		
Dose rate	7.42 kGy/h		
Date	2007/09/04		
Irradiation schedule			
	Sweep start	10:36	
	Irrad. start	10:56	
	Irrad. end	12:56	
	Sweep end	13:26	

Activity measurement

Run No.: 19-03 Date: 2007/09/04 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	07/09/04 10:42	300	40767	0.76	474.659	472.442	1.0000
Solution after irrad. 1mL	07/09/04 14:27	300	40857	0.80	475.707	479.922	9.76E-01
1: Ag-1	07/09/04 14:42	600	17877	1.32	278.054	280.770	5.94E-03
2: Ag-2	07/09/04 14:56	1800	6239	3.78	32.346	32.690	6.92E-04
3: Ag-3	07/09/04 16:00	1800	2464	8.69	12.775	12.960	2.74E-04
4: Ag-4	07/09/04 16:40	600	262	45.0	4.074	-	-
5: Ag-5	07/09/04 16:52	353	101	87.1	2.673	-	-
6: AgNO3-1	07/09/04 15:27	1800	8751	3.03	48.106	48.707	1.03E-03
7: AgNO3-2	07/09/04 17:00	303	-129	-	-	-	-
8: AgNO3-3	07/09/04 17:07	295	27	293	-	-	-

* Fraction to total inventory

Activity values for solution samples were for whole solution

Sum for filter 1-8	7.94E-03
Frac. in solution	9.76E-01
Loss	1.58E-02
Water loss by evaporation(%)	3.90

Chemical measurements

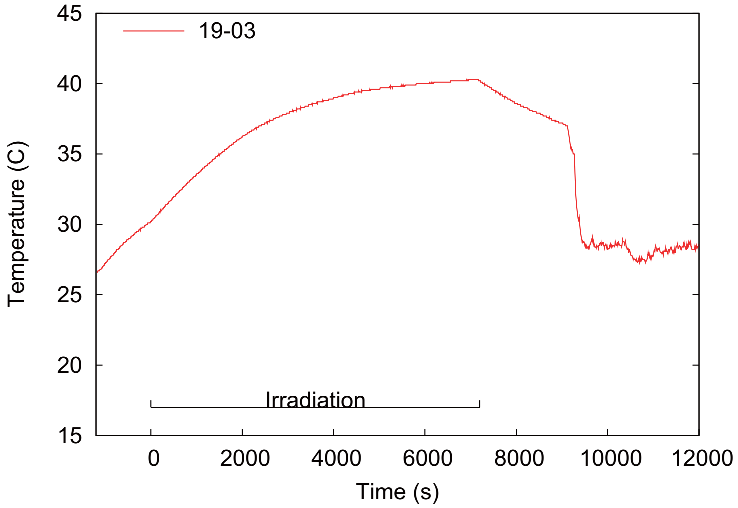
Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
19-03	8.412	8.388	9.98E-05	8.87E-05	8.89E-01

Filter fraction data

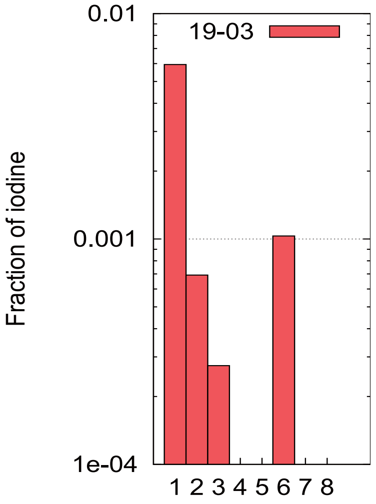
Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	5.943E-03	7.485E-01
2	6.919E-04	8.714E-02
3	2.743E-04	3.455E-02
4	-	-
5	-	-
AgNO3 (for Org-I)		
6	1.031E-03	1.298E-01
7	-	-
8	-	-

Plots

Temperature	Initial	27 ℃
	During irrad.	30--41 ℃

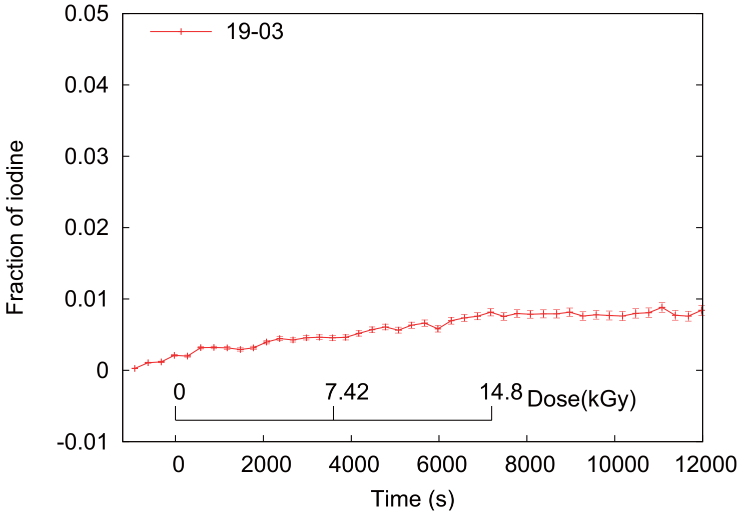


Distribution in filters



1-5: Ag filters
6-8: AgNO3 filters

Release history



Increasing trend before the start of irrad.
Suspected remnant of the previous run released from the pipe.
No increase after irradiation; the pipe was cleaned during this run?

Count efficiency(calibrated)= 0.1450

Experimental condition

Run No.	**19-03r**
Solution	pH=9.0 (Buffer: 0.1M B(OH)3-1.50e-5M NaOH), CsI 1e-4M, I-131 37kBq
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar
Gas	Air 1L/min
Dose rate	7.23 kGy/h
Date	2007/11/13

Irradiation schedule

Sweep start	13:18
Irrad. start	13:38
Irrad. end	15:38
Sweep end	16:08

Activity measurement

Run No.: 19-03r Date: 2007/11/13 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	07/11/13 13:30	1800.0	190006	0.37	370.712	373.056	1.0000
Solution after irrad. 1mL	07/11/13 16:18	1800.0	198241	0.36	386.775	393.164	1.042E+00
1: Ag-1	07/11/13 17:25	1800.0	15814	1.75	81.989	83.679	2.243E-03
2: Ag-2	07/11/13 18:42	323.8	71	122.54	2.046	-	-
3: Ag-3	07/11/13 18:49	410.9	90	104.44	2.044	-	-
4: Ag-4	07/11/13 18:57	305.4	72	109.7	2.200	-	-
5: Ag-5	07/11/13 19:04	302.1	12	633.3	0.371	-	-
6: AgNO3-1	07/11/13 18:02	1800.0	1427	15.21	7.844	8.024	2.151E-04
7: AgNO3-2	07/11/13 19:10	320.3	-29	-	0	-	-
8: AgNO3-3	07/11/13 19:17	313.6	-13	-	0	-	-

* Fraction to total inventory

Activity values for solution samples were for whole solution

Sum for filter 1-8	2.46E-03
Frac. in solution	1.04E+00
Loss	-4.48E-02
Water loss by evaporation(%)	1.10

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
19-03r	9.10	9.09	9.98E-05	1.50E-04	1.51E+00

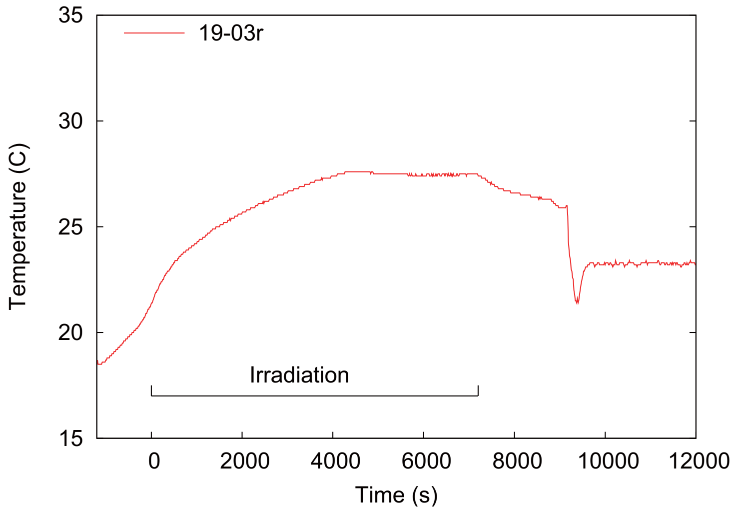
I- measurement problem?

Filter fraction data

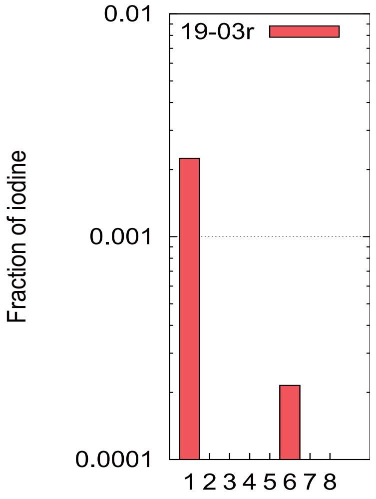
Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	2.243E-03	9.125E-01
2	-	-
3	-	-
4	-	-
5	-	-
AgNO3 (for Org-I)		
6	2.151E-04	8.750E-02
7	-	-
8	-	-

Plots

Temperature	Initial	18 ℃
	During irrad.	20--28 ℃

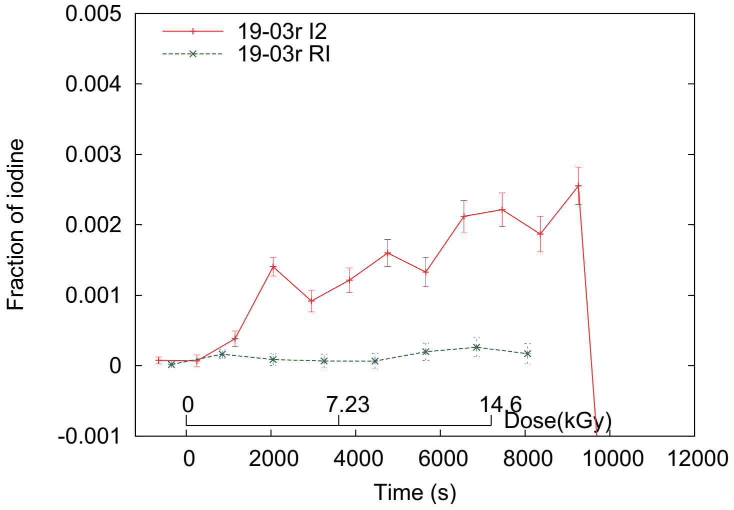


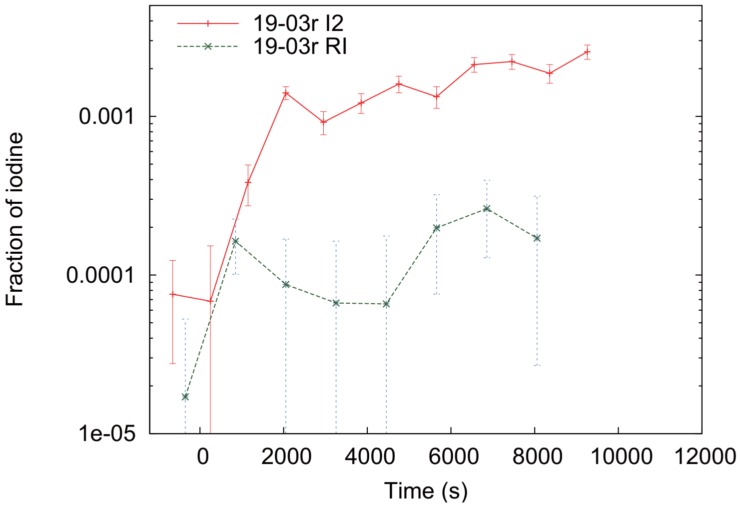
Distribution in filters



1-5: Ag filters
6-8: AgNO3 filters

Release history





Count efficiency(calibrated)= 0.1300 (Ag sampler)
0.1200 (AgNO3 sampler)

Experimental condition

Run No.	**19-04r**
Solution	pH=7.0 (Buffer: 0.1M B(OH)3-5.72e-4M NaOH), CsI 1e-4M, I-131 37kBq
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar
Gas	N2 1L/min
Dose rate	7.23 kGy/h
Date	2007/11/15
Irradiation schedule	

Sweep start	10:59
Irrad. start	11:39
Irrad. end	13:39
Sweep end	14:09

Activity measurement

Run No.: 19-04r Date: 2007/11/15 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	07/11/15 13:30	1800.0	196001	0.36	382.405	384.822	1.0000
Solution after irrad. 1mL	07/11/15 14:22	1800.0	106726	0.50	208.226	210.197	5.312E-01
1: Ag-1	07/11/15 15:14	404.1	667652	0.19	15419.422	15603.140	4.057E-01
2: Ag-2	07/11/15 15:23	604.9	231	49.35	3.564	3.609	9.382E-05
3: Ag-3	07/11/15 16:11	493.8	75	138.67	1.417	-	-
4: Ag-4	07/11/15 16:21	507.0	49	218.4	0.902	-	-
5: Ag-5	07/11/15 16:31	303.2	23	356.5	0.708	-	-
6: AgNO3-1	07/11/15 15:35	1094.2	11425	2.02	103.316	104.715	2.722E-03
7: AgNO3-2	07/11/15 15:54	472.6	-75	-	0	-	-
8: AgNO3-3	07/11/15 16:30	405.2	-95	-	0	-	-

* Fraction to total inventory

Activity values for solution samples were for whole solution

Sum for filter 1-8	4.08E-01
Frac. in solution	5.31E-01
Loss	6.03E-02

Sample mass(g)	Solution mass	Sample
Before	99.0	0.9905
After	97.7	1.0051

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
19-04r	6.72	6.77	9.98E-05	5.01E-05	5.02E-01

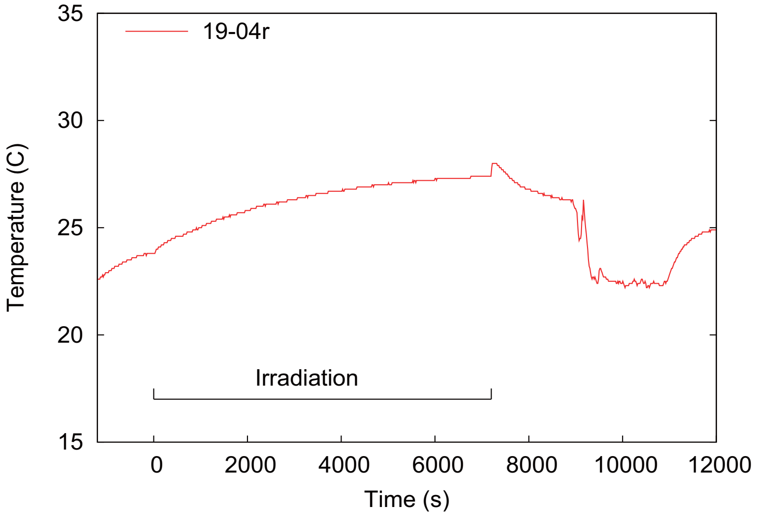
Filter fraction data

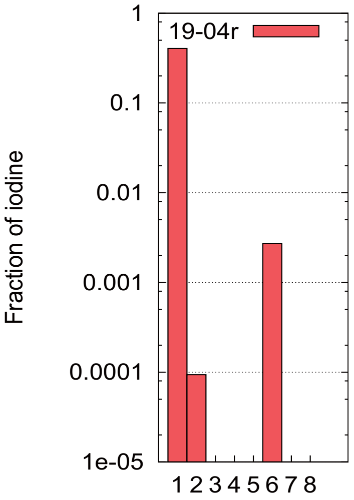
Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	4.057E-01	9.931E-01
2	9.382E-05	2.297E-04
3	-	-
4	-	-
5	-	-
AgNO3 (for Org-I)		
6	2.722E-03	6.665E-03
7	-	-
8	-	-

Plots

Temperature		Initial		23 ℃
			During irrad.	24--27 ℃

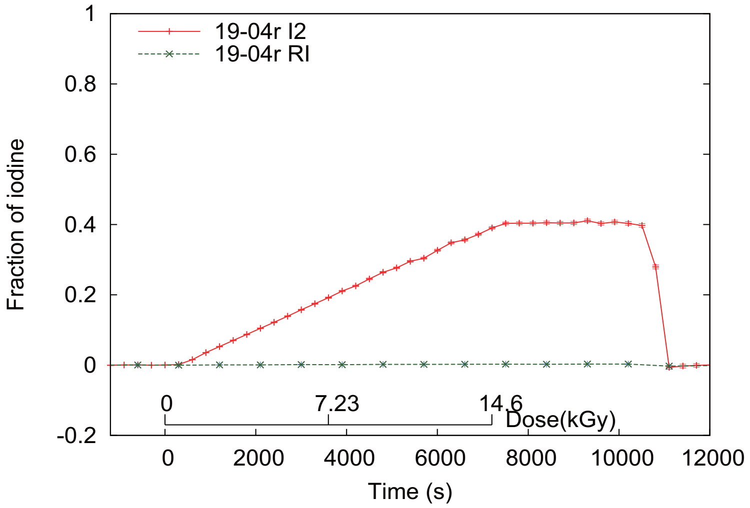
Distribution in filters

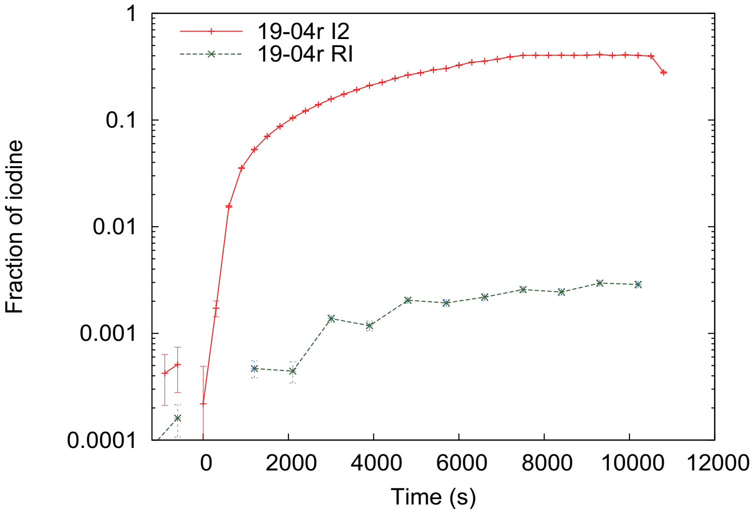




1-5: Ag filters
6-8: AgNO3 filters

Release history





Count efficiency(calibrated)=		0.1350 (Ag sampler)
					0.1500 (AgNO3 sampler)

Experimental condition

Run No.	**19-05**
Solution	pH=9.0 (Buffer: 0.1M B(OH)3-3.65e-2M NaOH), CsI 1e-4M, I-131 37kBq
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar
Gas	N2 1L/min
Dose rate	7.31 kGy/h
Date	2007/10/16

Irradiation schedule

Sweep start	10:38	
Irrad. start	10:58	
Irrad. end	14:58	*long irradiation: 4h
Sweep end	15:28	

Activity measurement

Run No.: 19-05 Date: 2007/10/16 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	07/10/17 09:55	486.8	41196	0.80	295.608	319.881	1.0000
Solution after irrad. 1mL	07/10/17 09:42	453.1	39598	0.81	305.235	330.042	9.740E-01
1: Ag-1	07/10/16 16:40	300.0	8738	1.93	271.799	276.404	8.641E-03
2: Ag-2	07/10/16 16:47	1444.7	5127	4.39	33.119	33.694	1.053E-03
3: Ag-3	07/10/16 18:10	1800.0	1520	14.41	7.881	8.057	2.519E-04
4: Ag-4	07/10/16 18:41	400.1	387	25.8	9.028	9.247	2.891E-04
5: Ag-5	07/10/16 18:49	449.1	97	112.4	2.015	-	-
6: AgNO3-1	07/10/16 17:13	1800.0	5510	4.39	30.289	30.863	9.648E-04
7: AgNO3-2	07/10/16 18:00	461.8	91	113.19	1.950	-	-
8: AgNO3-3	07/10/16 17:52	311.5	-153	-	0	-	-

* Fraction to total inventory
Activity values for solution samples were for whole solution

Sum for filter 1-8	1.12E-02
Frac. in solution	9.74E-01
Loss	1.48E-02
Water loss by evaporation(%)	5.60

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
19-05	9.125	9.129	9.98E-05	6.99E-05	7.01E-01

Filter fraction data

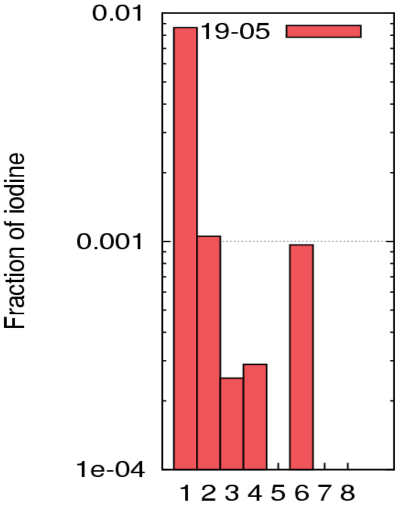
Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	8.641E-03	7.715E-01
2	1.053E-03	9.405E-02
3	2.519E-04	2.249E-02
4	2.891E-04	2.581E-02
5	-	-
AgNO3 (for Org-I)		
6	9.648E-04	8.615E-02
7	-	-
8	-	-

Plots

Temperature	Initial	約25℃
	During irrad.	約38℃

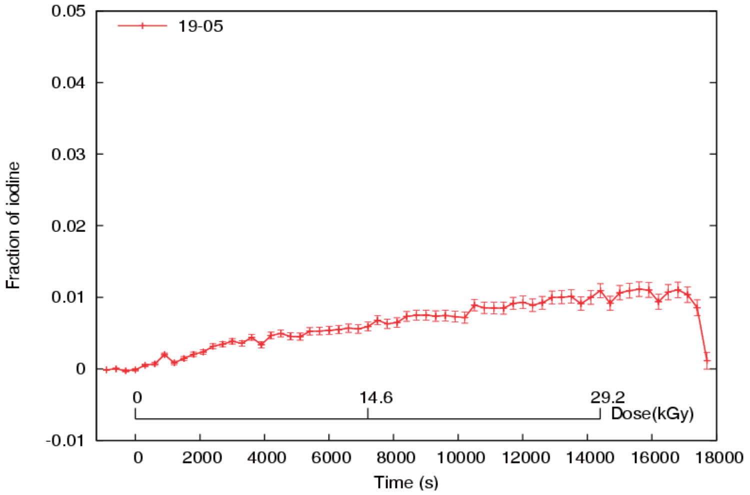
* Temperature data retrieval failed.

Distribution in filters



1-5: Ag filters
6-8: AgNO3 filters

Release history



Count efficiency(calibrated)= 0.1600

Experimental condition

Run No.	**19-06m**
Solution	pH=7.0 (Buffer: 0.1M B(OH)3-5.72e-4M NaOH), CsI 1e-4M, Fe^2+ 1e-4M, I-131 37kBq
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar
Gas	Air 1L/min
Dose rate	7.219 kGy/h
Date	2007/11/20

*Fe^2+: FeSO4 7H2O 0.1Maq 0.121mL added to the test solution 121mL

Irradiation schedule

Sweep start	11:50
Irrad. start	12:21
Irrad. end	14:21
Sweep end	14:51

Activity measurement

Run No.: 19-06m Date: 2007/11/20 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	07/11/20 11:43	1800.0	200228	3.6E-1	390.656	390.609	1.0000
Solution after irrad. 1mL	07/11/20 15:05	1800.0	175503	3.8E-1	342.416	346.551	8.666E-01
1: Ag-1	07/11/20 15:52	509.6	206974	3.3E-1	3790.432	3844.554	9.900E-02
2: Ag-2	07/11/20 16:03	613.8	29220	9.8E-1	444.276	450.941	1.161E-02
3: Ag-3	07/11/20 16:14	302.4	111	7.3E+1	3.426	-	-
4: Ag-4	07/11/20 16:54	376.1	32	2.8E+2	0.794	-	-
5: Ag-5	07/11/20 17:01	384.8	32	2.9E+2	0.776	-	-
6: AgNO3-1	07/11/20 16:21	1800.0	1642	1.3E+1	9.026	9.177	2.363E-04
7: AgNO3-2	07/11/20 17:09	327.1	49	1.7E+2	1.482	-	-
8: AgNO3-3	07/11/20 17:16	302.9	52	1.5E+2	1.699	-	-

Sum for filter 1-8	1.11E-01
Frac. in solution	8.67E-01
Loss	2.25E-02

* Fraction to total inventory

Activity values for solution samples were for whole solution

Sample mass(g)	Solution mass	Sample
Before	98.7	0.9928
After	97.2	1.0010

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
19-06m	6.85	6.73	9.98E-05	8.11E-05	8.12E-01

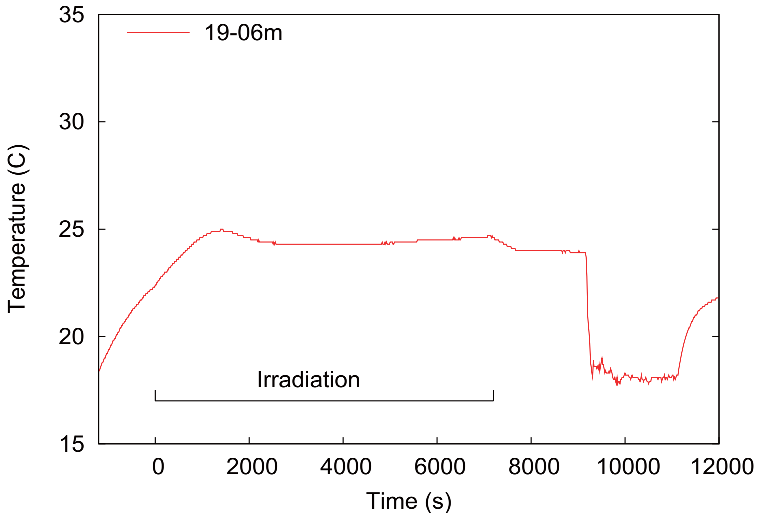
Filter fraction data

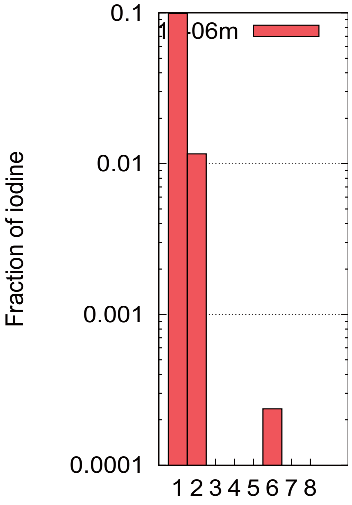
Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	9.900E-02	8.931E-01
2	1.161E-02	1.048E-01
3	-	-
4	-	-
5	-	-
AgNO3 (for Org-I)		
6	2.363E-04	2.132E-03
7	-	-
8	-	-

Plots

Temperature	Initial	18 ℃
	During irrad.	22--25 ℃

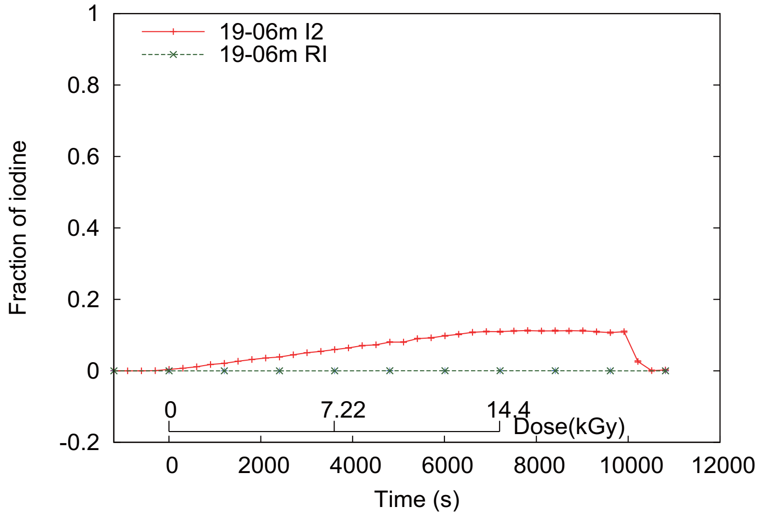
Distribution in filters

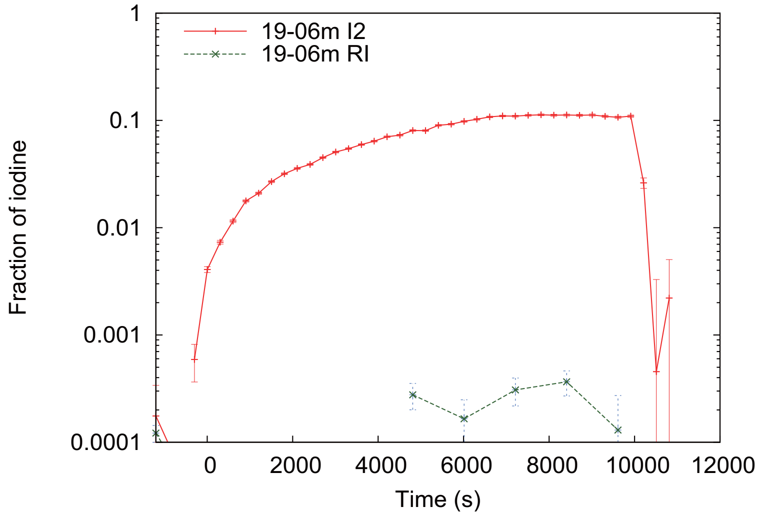




1-5: Ag filters
6-8: AgNO3 filters

Release history





Count efficiency(calibrated)= 0.1320 (Ag sampler)
0.1500 (AgNO3 sampler)

Experimental condition

Run No.	**19-07m**
Solution	pH=7.0 (Buffer: 0.1M B(OH)3-5.72e-4M NaOH), CsI 1e-4M, MIBK 1e-4M, I-131 37kBq)
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar
Gas	Air 1L/min
Dose rate	7.19 kGy/h
Date	2007/11/30

Irradiation schedule

Sweep start	11:01
Irrad. start	11:31
Irrad. end	13:31
Sweep end	14:00

Activity measurement

Run No.: 19-07m Date: 2007/11/30 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	07/11/30 11:07	1800.0	205681	3.6E-1	401.295	400.381	1.0000
Solution after irrad. 1mL	07/11/30 14:12	1800.0	190779	3.7E-1	372.216	375.514	9.126E-01
1: Ag-1	07/11/30 15:14	531.2	189319	3.4E-1	3325.994	3365.836	8.333E-02
2: Ag-2	07/11/30 15:24	1235.9	1379	1.2E+1	10.413	10.548	2.611E-04
3: Ag-3	07/11/30 16:21	175.6	22	3.0E+2	1.169	-	-
4: Ag-4	07/11/30 16:25	151.7	-4	-	0	-	-
5: Ag-5	07/11/30 16:29	152.7	83	6.7E+1	5.071	-	-
6: AgNO3-1	07/11/30 15:47	1800.0	4070	5.3E+0	22.373	22.701	5.620E-04
7: AgNO3-2	07/11/30 16:32	151.2	-42	-	0	-	-
8: AgNO3-3	07/11/30 16:37	152.3	-18	-	0	-	-

Sum for filter 1-8	8.41E-02
Frac. in solution	9.13E-01
Loss	3.23E-03

* Fraction to total inventory

Activity values for solution samples were for whole solution

Sample mass(g)	Solution mass	Sample
Before	100.1	0.9922
After	98.6	1.0044

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
19-07m	6.99	6.92	9.98E-05	7.98E-05	7.99E-01

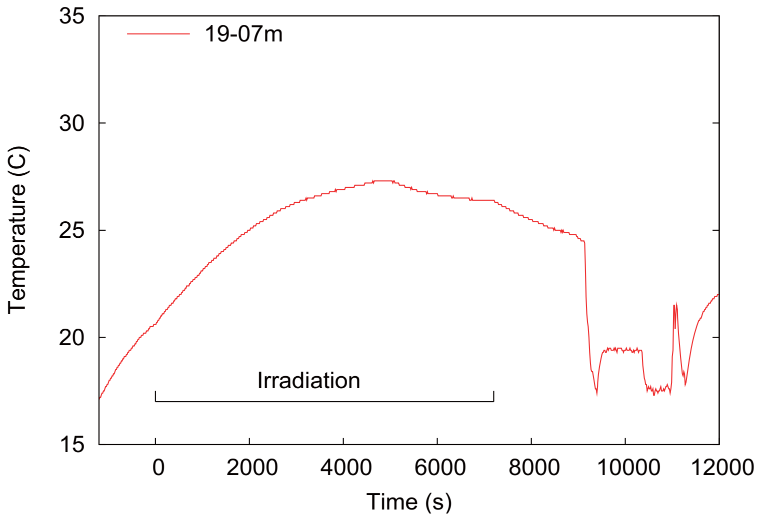
Filter fraction data

Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	8.333E-02	9.902E-01
2	2.611E-04	3.103E-03
3	-	-
4	-	-
5	-	-
AgNO3 (for Org-I)		
6	5.620E-04	6.678E-03
7	-	-
8	-	-

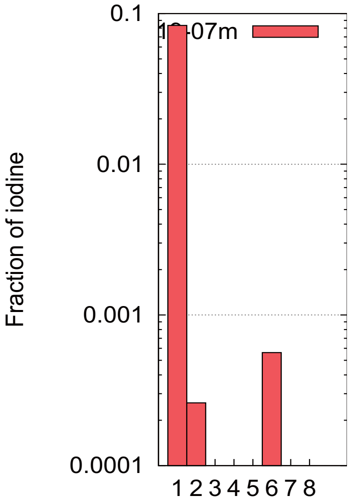
No significant adsorption in the up-stream coupler of the top sampler (as in 19-08m).
A temperature control mistake caused too high temperatures. No troubles such as condensation in the samplers.

Plots

Temperature	Initial	17 ℃
	During irrad.	21--27 ℃

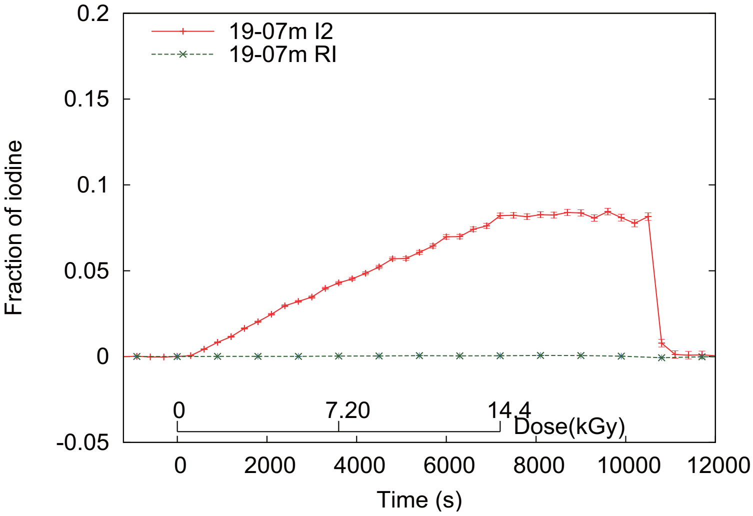


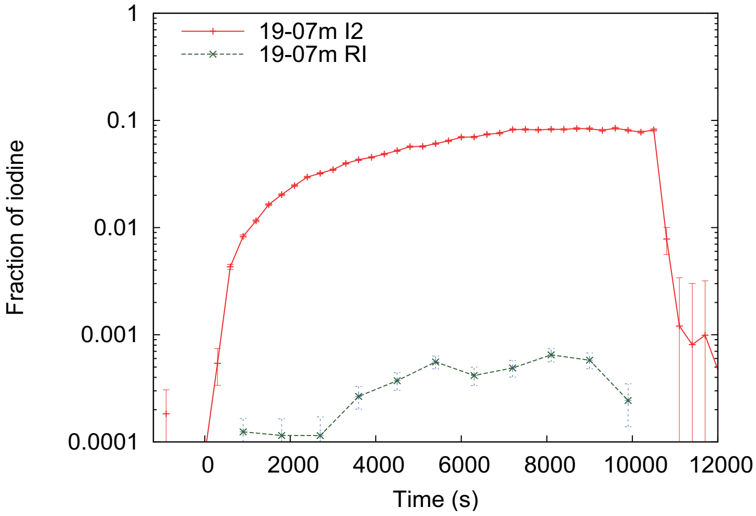
Distribution in filters



1-5: Ag filters
6-8: AgNO3 filters

Release history





Count efficiency(calibrated)= 0.1590 (Ag sampler)
0.2000 (AgNO3 sampler)

Experimental condition

Run No.	**19-08m**	
Solution	pH=7.0 (Buffer: 0.1M B(OH)3-5.72e-4M NaOH), CsI 1e-4M, I-131 37kBq; 照射直前に0.05M MIBK水溶液0.2mL添加(1e-4M)	
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar	
Gas	N2 1L/min	
Dose rate	7.20 kGy/h	
Date	2007/11/27	
Irradiation schedule		
	Sweep start	10:56
	Irrad. start	11:27
	Irrad. end	13:27
	Sweep end	13:57

Activity measurement

Run No.: 19-08m Date: 2007/11/27 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	07/11/27 11:00	1800.0	187395	3.7E-1	365.618	364.632	1.0000
Solution after irrad. 1mL	07/11/27 14:14	1800.0	150803	4.2E-1	294.222	296.864	8.088E-01
1: Ag-1	07/11/27 15:33	401.6	181067	3.5E-1	4207.775	4262.762	1.164E-01
2: Ag-2	07/11/27 15:42	1012.2	1636	9.9E+0	15.084	15.294	4.175E-04
3: Ag-3	07/11/27 16:33	605.1	511	2.4E+1	7.881	8.013	2.187E-04
4: Ag-4	07/11/27 16:44	655.4	468	2.6E+1	6.664	6.781	1.851E-04
5: Ag-5	07/11/27 16:57	336.6	110	8.2E+1	3.050	-	-
6: AgNO3-1	07/11/27 16:01	1800.0	35706	1.0E+0	196.281	199.320	5.440E-03
7: AgNO3-2	07/11/27 17:04	152.0	-70	-	0	-	-
8: AgNO3-3	07/11/27 17:08	150.5	-26	-	0	-	-

Sum for filter 1-8	1.23E-01
Frac. in solution	8.09E-01
Loss	6.86E-02

* Fraction to total inventory

Activity values for solution samples were for whole solution

Sample mass(g)	Solution mass	Sample
Before	99.7	0.9923
After	98.4	0.9859

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
19-08m	6.99	6.96	9.98E-05	6.52E-05	6.54E-01

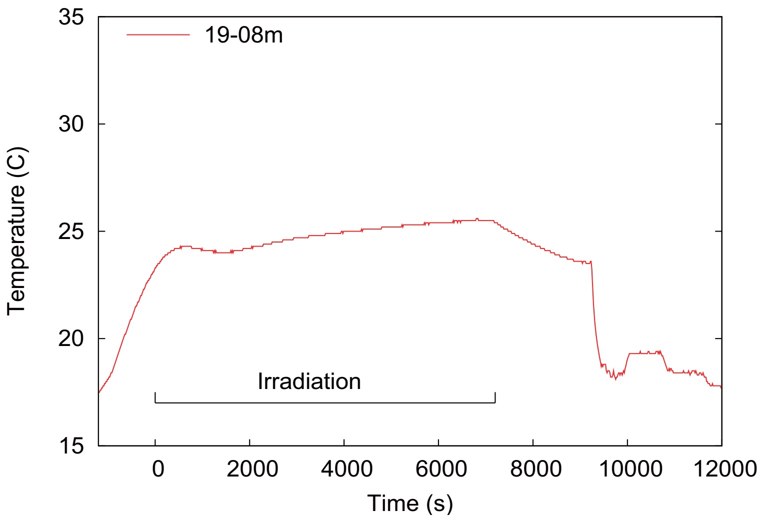
Filter fraction data

Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	1.164E-01	9.489E-01
2	4.175E-04	3.405E-03
3	2.187E-04	1.784E-03
4	1.851E-04	1.509E-03
5	-	-
AgNO3 (for Org-I)		
6	5.440E-03	4.437E-02
7	-	-
8	-	-

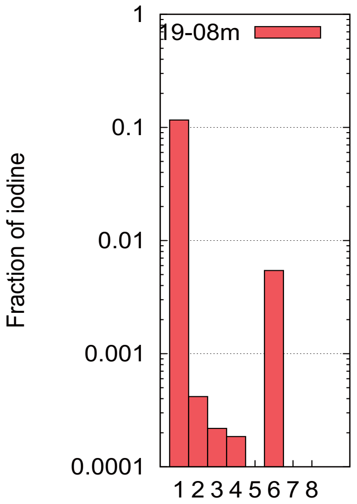
A significant adsorption was indicated at the up-stream coupler of the top sampler. 480cps(>1.5kBq)
Large loss, 6.9%, compared to the release, 12%. A large part of the loss should be in the coupler.

Plots

Temperature Initial 17 ℃
During irrad. 23--26 ℃

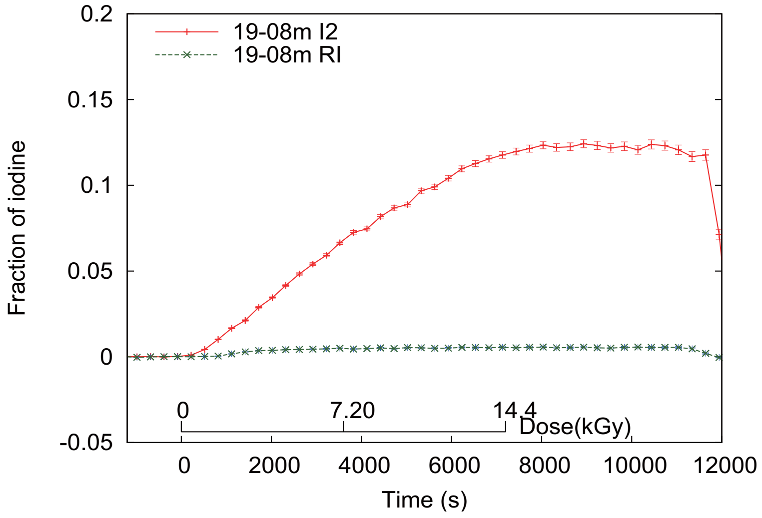


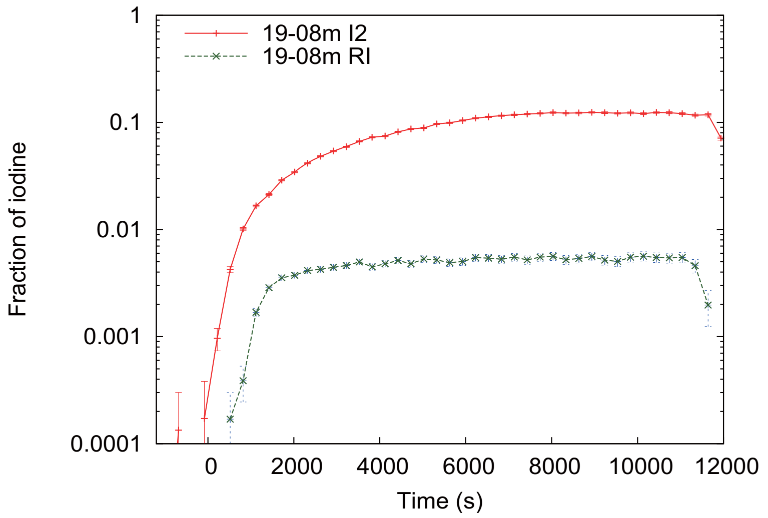
Distribution in filters



1-5: Ag filters
6-8: AgNO3 filters

Release history





Count efficiency(calibrated)= 0.1620 (Ag sampler)
0.2050 (AgNO3 sampler)

Tapered PTFE rings were inserted at the entrance of samplers; that probably enhanced the efficiency of the detection.

Experimental condition

Run No.	**19-09**	
Solution	pH=7.0 (Buffer: 0.1M B(OH)3-5.72e-4M NaOH), CsI 1e-4M, I-131 37kBq; NaCl 1e-3M	
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar	
Gas	Air 1L/min	
Dose rate	7.00 kGy/h	
Date	2008/02/14	
Irradiation schedule		
	Sweep start	11:03
	Irrad. start	11:33
	Irrad. end	13:33
	Sweep end	14:03

Activity measurement

Run No.: 19-09 Date: 2008/02/14 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	08/02/14 17:27	1800.0	186587	3.7E-1	364.037	371.586	1.0000
Solution after irrad. 1mL	08/02/14 14:22	1800.0	170651	3.9E-1	332.946	336.098	8.989E-01
1: Ag-1	08/02/14 15:12	337.2	142752	3.9E-1	3951.222	3997.686	1.067E-01
2: Ag-2	08/02/14 15:22	1800.0	3909	5.4E+0	20.266	20.532	5.480E-04
3: Ag-3	08/02/14 16:37	1473.2	1160	1.5E+1	7.348	7.477	1.996E-04
4: Ag-4	08/02/14 17:03	600.0	203	5.2E+1	3.157	-	-
5: Ag-5	08/02/14 17:14	154.2	115	4.6E+1	6.959	-	-
6: AgNO3-1	08/02/14 16:04	1800.0	4113	5.4E+0	22.610	22.964	6.130E-04
7: AgNO3-2	08/02/14 17:18	156.2	-24	-	0	-	-
8: AgNO3-3	08/02/14 17:23	153.9	-40	-	0	-	-

Sum for filter 1-8	1.08E-01
Frac. in solution	8.99E-01
Loss	-6.94E-03

* Fraction to total inventory

Activity values for solution samples were for whole solution

Sample mass(g)	Solution mass	Sample
Before	101.4	1.0057
After	100.1	0.9991

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
19-09	7.03	7.00	9.96E-05	9.30E-05	9.33E-01

Filter fraction data

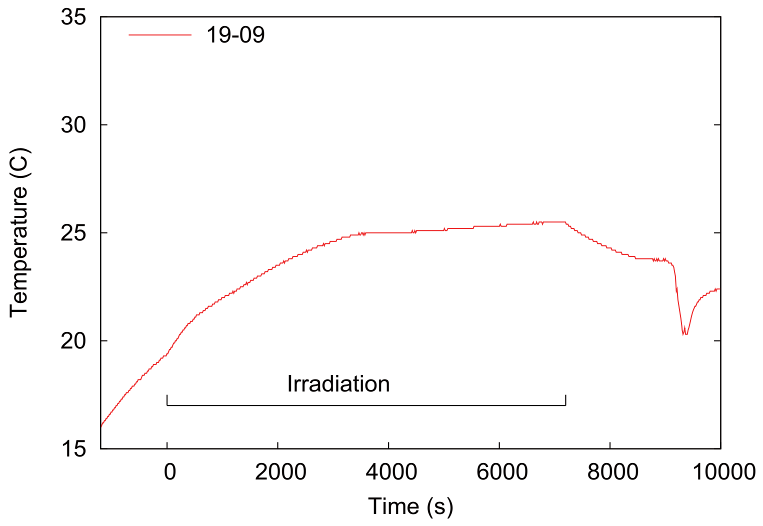
Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	1.067E-01	9.874E-01
2	5.480E-04	5.071E-03
3	1.996E-04	1.847E-03
4	-	-
5	-	-
AgNO3 (for Org-I)		
6	6.130E-04	5.672E-03
7	-	-
8	-	-

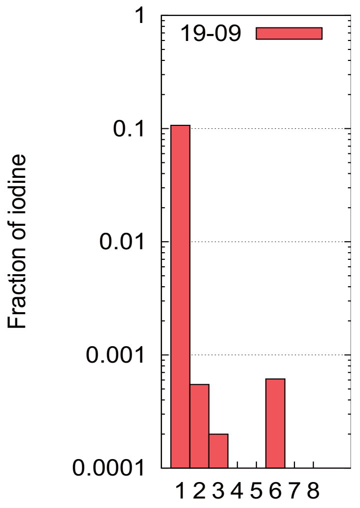
Plots

Temperature Initial 17 ℃

During irrad. 20--26 ℃

Distribution in filters

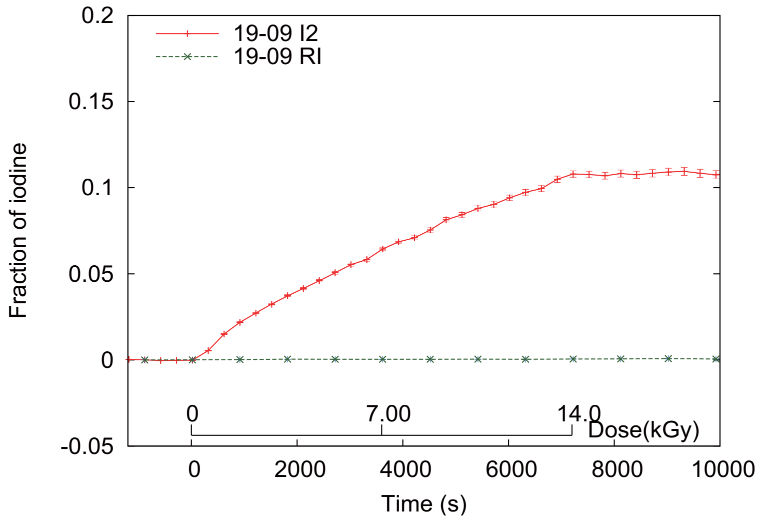


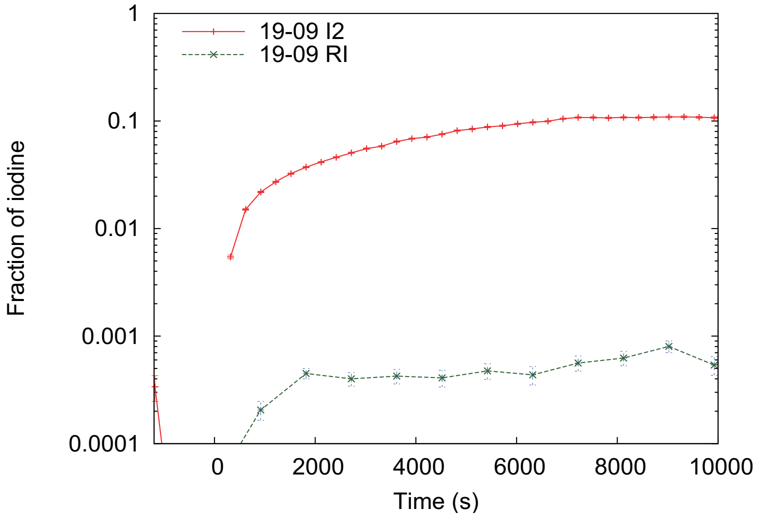


1-5: Ag filters

6-8: AgNO3 filters

Release history





Count efficiency(calibrated)= 0.1650 (Ag sampler)

0.2050 (AgNO3 sampler)

Experimental condition

Run No.	**20-01**	
Solution	pH=7.0 (Buffer: 0.1M B(OH)3-5.72e-4M NaOH), CsI 1e-4M, I-131 37kBq; MIBK 1e-3M(added at the start of irrad.)	
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar	
Gas	Air 1L/min	
Dose rate	6.91 kGy/h	
Date	2008/06/18	
Irradiation schedule		
	Sweep start	11:14
	Irrad. start	11:44
	Irrad. end	13:44
	Sweep end	14:14

Activity measurement

Run No.: 20-01 Date: 2008/06/18 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	08/06/18 11:18	1800.0	358768	2.6E-1	699.968	698.835	1.0000
Solution after irrad. 1mL	08/06/18 15:20	1800.0	303076	2.8E-1	591.318	598.997	8.652E-01
1: Ag-1	08/06/18 16:03	314.1	211291	3.4E-1	6278.068	6371.291	9.163E-02
2: Ag-2	08/06/18 16:10	637.6	3697	4.1E+0	54.109	54.945	7.902E-04
3: Ag-3	08/06/18 16:49	300.0	1167	6.9E+0	36.300	36.941	5.313E-04
4: Ag-4	08/06/18 16:57	300.0	360	2.3E+1	11.198	11.401	1.640E-04
5: Ag-5	08/06/18 17:04	300.0	529	1.8E+1	16.456	16.761	2.411E-04
6: AgNO3-1	08/06/18 16:25	1327.7	37405	9.8E-1	278.770	283.427	4.076E-03
7: AgNO3-2	08/06/18 17:12	210.0	36	1.7E+2	1.696	-	-
8: AgNO3-3	08/06/18 17:17	157.9	-107	-	0	-	-

* Fraction to total inventory

Activity values for solution samples were for whole solution

Sum for filter 1-8	9.74E-02
Frac. in solution	8.65E-01
Loss	3.74E-02

Sample mass(g)	Solution mass	Sample
Before	100.1	1.0061
After	100.7	1.0027

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
20-01	6.93	6.37	9.98E-05	8.06E-05	8.08E-01

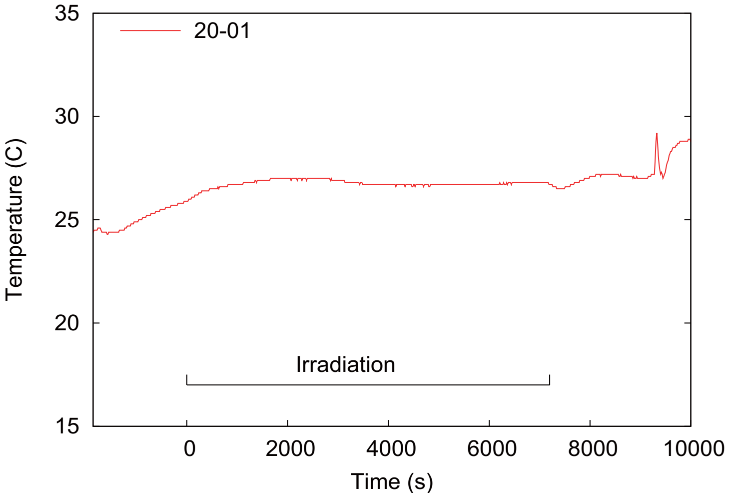
Filter fraction data

Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	9.163E-02	9.404E-01
2	7.902E-04	8.110E-03
3	5.313E-04	5.453E-03
4	1.640E-04	1.683E-03
5	2.411E-04	2.474E-03
AgNO3 (for Org-I)		
6	4.076E-03	4.184E-02
7	-	-
8	-	-

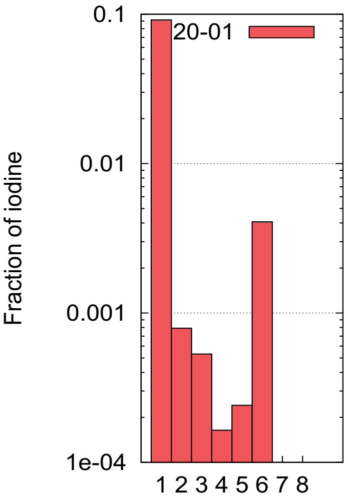
Adsorption was found at the up-stream coupler of the top sampler: 80cps(about 300Bq); Loss was relatively small.

Plots

Temperature	Initial	24 C
	During irrad.	25--27 C

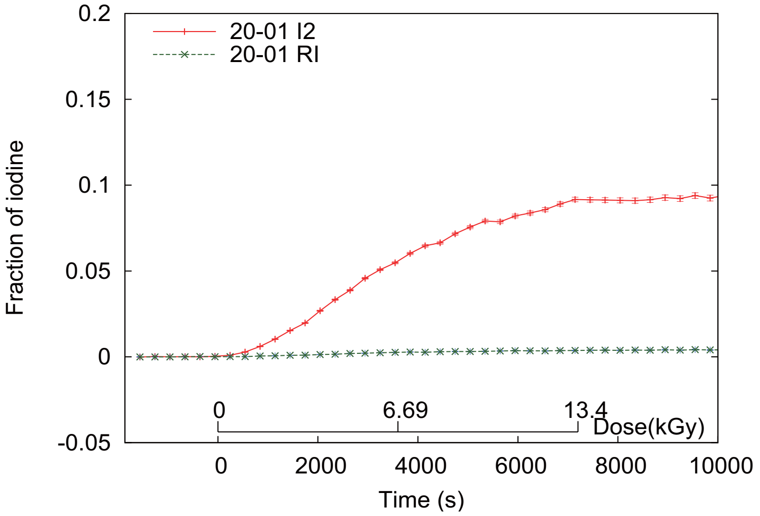


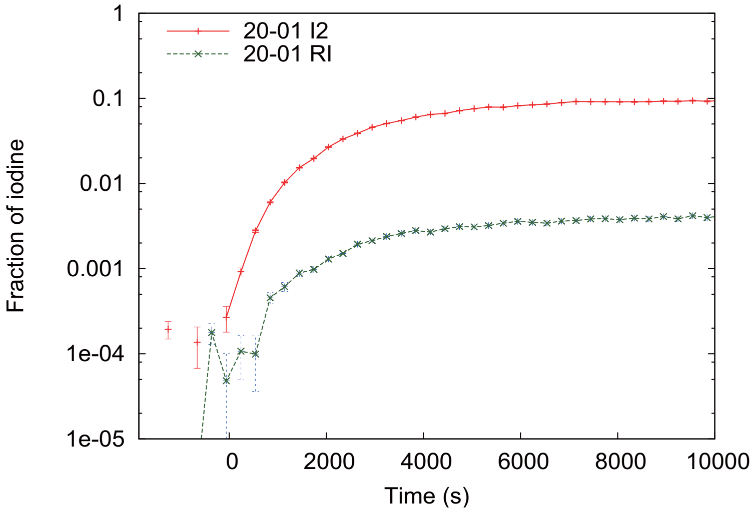
Distribution in filters



1-5: Ag filters
6-8: AgNO3 filters

Release history





Count efficiency(calibrated)= 0.1580 (Ag sampler)
0.2200 (AgNO3 sampler)

Tapered PTFE rings were inserted at the inlet of samplers.

Experimental condition

Run No.	**20-02**
Solution	pH=7.0 (Buffer: 0.1M B(OH)3-5.72e-4M NaOH), CsI 1e-4M, I-131 37kBq; MIBK 1e-3M(added at the start of irrad.)
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar
Gas	N2 1L/min
Dose rate	6.89 kGy/h
Date	2008/06/25

Irradiation schedule

Sweep start	10:47	
Irrad. start	11:17	
Irrad. end	13:17	
Sweep end	14:07	The filter activity continued to increase after the irrad. end; the sweep was extended a till flat trend was confirmed.

Activity measurement

Run No.: 20-02 Date: 2008/06/25 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	08/06/25 10:51	1789.4	194845	3.6E-1	382.405	381.166	1.0000
Solution after irrad. 1mL	08/06/25 15:24	1800.0	184981	3.6E-1	360.904	365.679	9.768E-01
1: Ag-1	08/06/25 16:10	609.4	30600	9.4E-1	468.618	475.846	1.265E-02
2: Ag-2	08/06/25 16:36	600.0	1508	8.3E+0	23.454	23.853	6.339E-04
3: Ag-3	08/06/25 16:55	600.0	970	1.2E+1	15.087	15.361	4.082E-04
4: Ag-4	08/06/25 17:11	152.1	34	1.7E+2	2.086	-	-
5: Ag-5	08/06/25 17:15	150.5	-28	-	0	-	-
6: AgNO3-1	08/06/25 16:21	600.0	18320	1.4E+0	302.113	306.974	8.158E-03
7: AgNO3-2	08/06/25 16:51	152.2	-57	-	0	-	-
8: AgNO3-3	08/06/25 17:19	160.1	-11	-	0	-	-

Sum for filter 1-8	2.18E-02
Frac. in solution	9.77E-01
Loss	1.34E-03

* Fraction to total inventory

Activity values for solution samples were for whole solution

Sample mass(g)	Solution mass	Sample
Before	99.3	1.0059
After	99.9	0.9939

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
20-02	7.16	7.10	9.98E-05	8.38E-05	8.40E-01

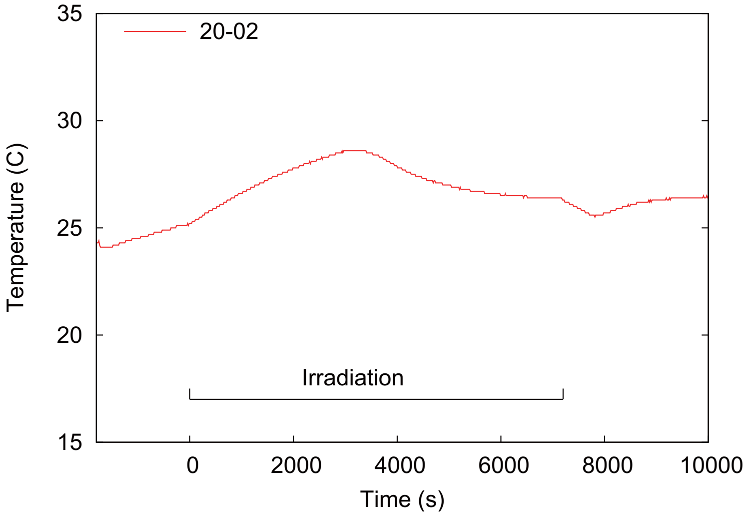
Filter fraction data

Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	1.265E-02	5.789E-01
2	6.339E-04	2.902E-02
3	4.082E-04	1.869E-02
4	-	-
5	-	-
AgNO3 (for Org-I)		
6	8.158E-03	3.734E-01
7	-	-
8	-	-

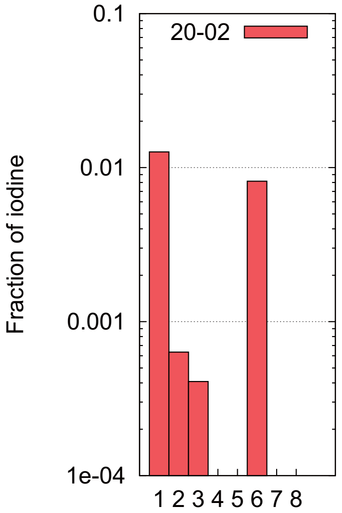
Adsorption was found at the up-stream coupler of the top sampler: 13cps(about 50Bq); Loss was small.

Plots

Temperature Initial 24 C
During irrad. 25--29 C

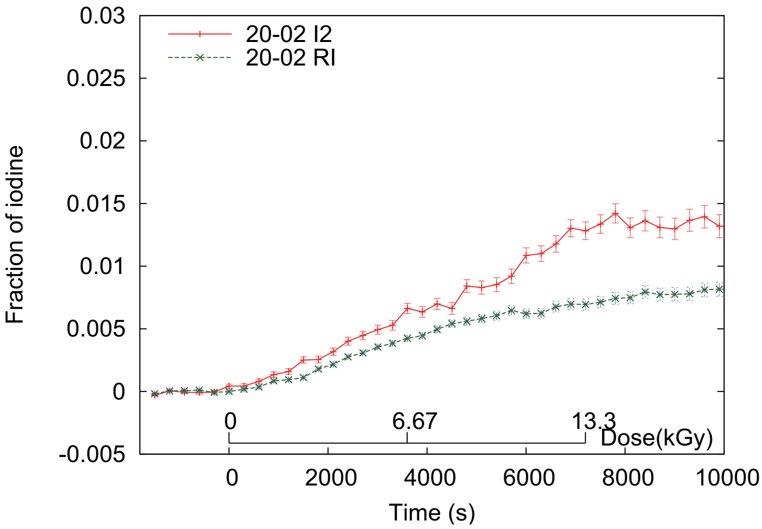


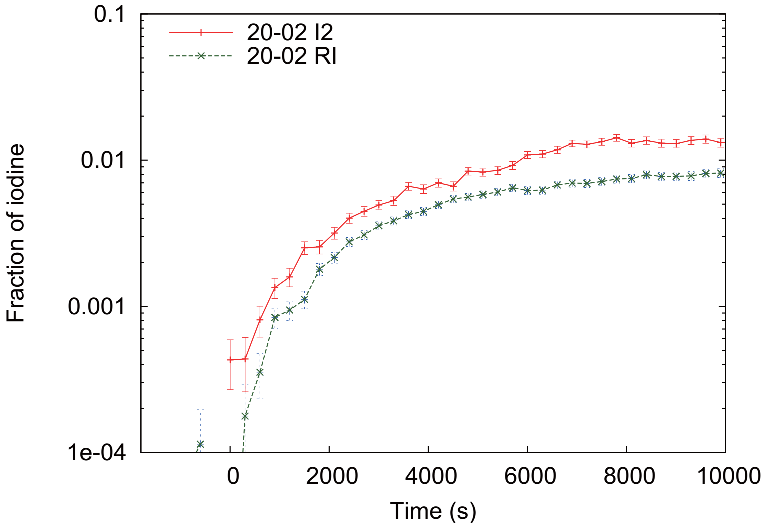
Distribution in filters



1-5: Ag filters
6-8: AgNO3 filters

Release history





Count efficiency(calibrated)= 0.1620 (Ag sampler)
0.2030 (AgNO3 sampler)

Tapered PTFE rings were inserted at the inlet of samplers.

Experimental condition

Run No.	**20-03**
Solution	pH=7.0 (Buffer: 0.1M B(OH)3-5.72e-4M NaOH), CsI 1e-4M, I-131 37kBq; MIBK 1e-3M(added at the start of irrad.)
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar
Gas	Ar 1L/min (mass flow controller correction factor=1.4, setting=0.714SLM)
Dose rate	6.87 kGy/h
Date	2008/07/03

Irradiation schedule

Sweep start	12:08
Irrad. start	12:38
Irrad. end	14:38
Sweep end	15:08

Activity measurement

Run No.: 20-03 Date: 2008/07/03 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	08/07/03 12:26	1800.0	188735	3.7E-1	368.232	369.140	1.0000
Solution after irrad. 1mL	08/07/03 16:16	1800.0	180114	3.7E-1	351.412	357.174	9.798E-01
1: Ag-1	08/07/03 16:52	1201.6	48811	7.9E-1	379.084	386.018	1.055E-02
2: Ag-2	08/07/03 17:44	604.2	626	2.0E+1	9.669	9.874	2.700E-04
3: Ag-3	08/07/03 17:55	601.0	552	1.9E+1	8.572	8.759	2.395E-04
4: Ag-4	08/07/03 18:09	317.2	195	4.2E+1	5.737	5.867	1.604E-04
5: Ag-5	08/07/03 18:16	150.9	78	6.5E+1	4.823	-	-
6: AgNO3-1	08/07/03 17:13	1800.0	35340	1.0E+0	194.269	198.131	5.417E-03
7: AgNO3-2	08/07/03 18:19	234.1	97	6.2E+1	4.101	-	-
8: AgNO3-3	08/07/03 18:24	152.9	6	8.5E+2	0.388	-	-

Sum for filter 1-8	1.66E-02
Frac. in solution	9.80E-01
Loss	3.53E-03

* Fraction to total inventory

Activity values for solution samples were for whole solution

Sample mass(g)	Solution mass	Sample
Before	99.6	1.0053
After	100.7	1.0037

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
20-03	6.98	6.96	9.98E-05	8.78E-05	8.80E-01

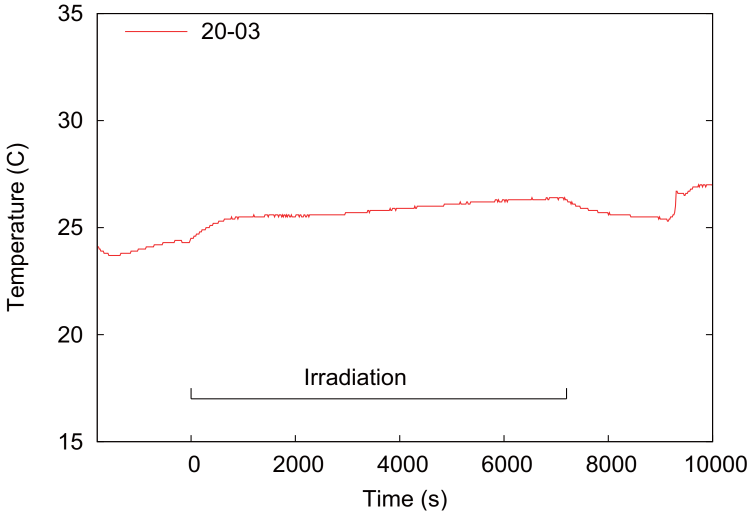
Filter fraction data

Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	1.055E-02	6.342E-01
2	2.700E-04	1.622E-02
3	2.395E-04	1.439E-02
4	1.604E-04	9.639E-03
5	-	-
AgNO3 (for Org-I)		
6	5.417E-03	3.255E-01
7	-	-
8	-	-

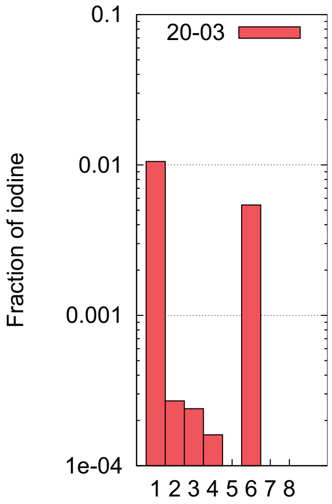
Adsorption was found at the up-stream coupler of the top sampler: 13cps(about 50Bq); Loss was small.
A slight white suspension was observed after irrad.
(not checked in the case with N2 gas)

Plots

Temperature	Initial	24 C
	During irrad.	24--26 C

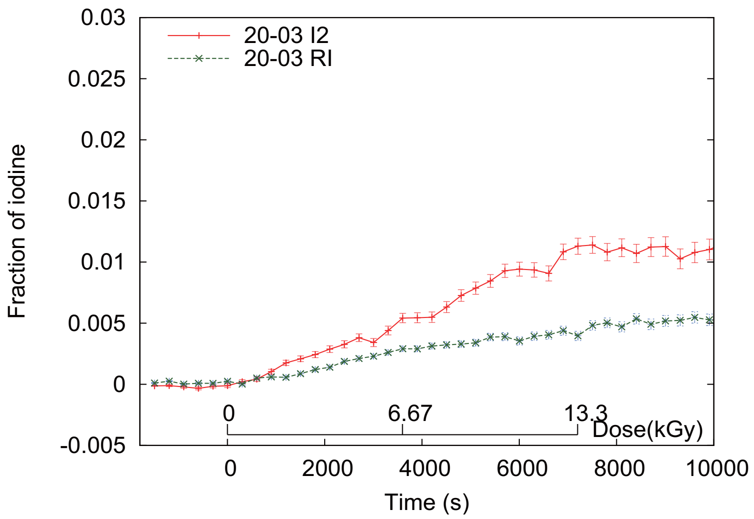


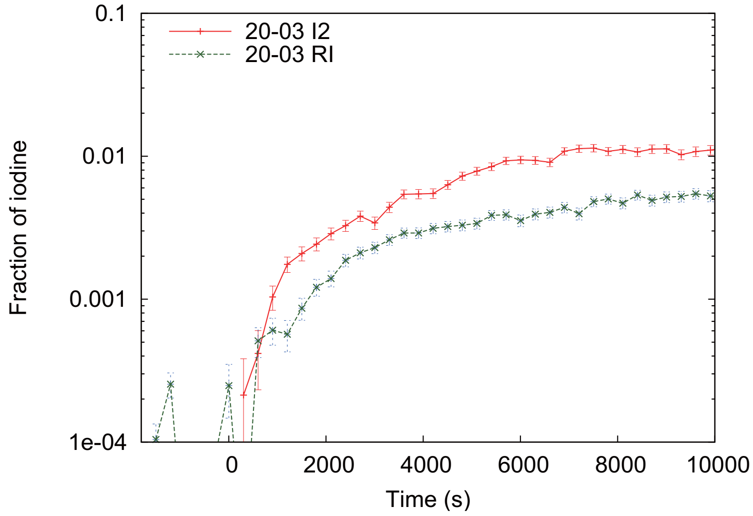
Distribution in filters



1-5: Ag filters
6-8: AgNO3 filters

Release history





Count efficiency(calibrated)= 0.1640 (Ag sampler)
0.2080 (AgNO3 sampler)

Tapered PTFE rings were inserted at the inlet of samplers.

Experimental condition

Run No.	**20-04**	
Solution	pH=7.0 (Buffer: 0.1M B(OH)3-5.72e-4M NaOH), CsI 1e-4M, I-131 37kBq	
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar	
Temperature	40C	
Gas	Air 1L/min	
Dose rate	6.79 kGy/h	
Date	2008/08/07	
Irradiation schedule		
	Sweep start	10:48
	Irrad. start	11:18
	Irrad. end	13:18
	Sweep end	13:48

Activity measurement

Run No.: 20-04 Date: 2008/08/07 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	08/08/07 10:58	1800.0	228025	3.3E-1	444.889	443.636	1.0000
Solution after irrad. 1mL	08/08/07 15:08	1800.0	216177	3.4E-1	421.773	426.943	9.301E-01
1: Ag-1	08/08/07 15:40	600.6	101370	5.2E-1	1575.210	1596.624	3.655E-02
2: Ag-2	08/08/07 16:24	600.0	25368	1.1E+0	394.567	400.988	9.179E-03
3: Ag-3	08/08/07 16:37	600.0	947	1.4E+1	14.729	14.981	3.429E-04
4: Ag-4	08/08/07 16:48	600.0	481	2.2E+1	7.481	7.614	1.743E-04
5: Ag-5	08/08/07 17:00	151.7	-93	-	0	-	-
6: AgNO3-1	08/08/07 15:52	1800.0	3053	7.0E+0	16.783	17.033	3.899E-04
7: AgNO3-2	08/08/07 17:03	151.5	27	2.0E+2	1.763	-	-
8: AgNO3-3	08/08/07 17:07	151.5	32	1.8E+2	2.090	-	-

* Fraction to total inventory
Activity values for solution samples were for whole solution

Sum for filter 1-8	4.66E-02
Frac. in solution	9.30E-01
Loss	2.32E-02

Sample mass(g)	Solution mass	Sample
Before	98.9	1.0044
After	94.9	0.9972

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
20-04	7.01	6.98	9.98E-05	9.34E-05	9.36E-01

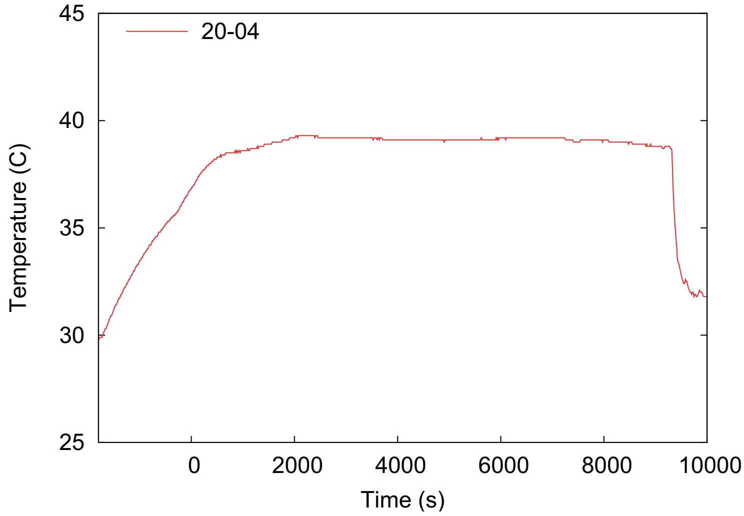
Filter fraction data

Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	3.655E-02	7.837E-01
2	9.179E-03	1.968E-01
3	3.429E-04	7.353E-03
4	1.743E-04	3.737E-03
5	-	-
AgNO3 (for Org-I)		
6	3.899E-04	8.361E-03
7	-	-
8	-	-

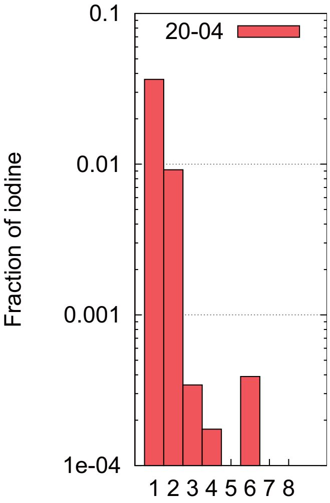
Adsorption was found at the up-stream coupler of the top sampler: 48cps(about 170Bq), PTFE ring: 3cps(about 20Bq)
Condensation in the top sampler.

Plots

Temperature	Initial	30 C
	During irrad.	38--39 C

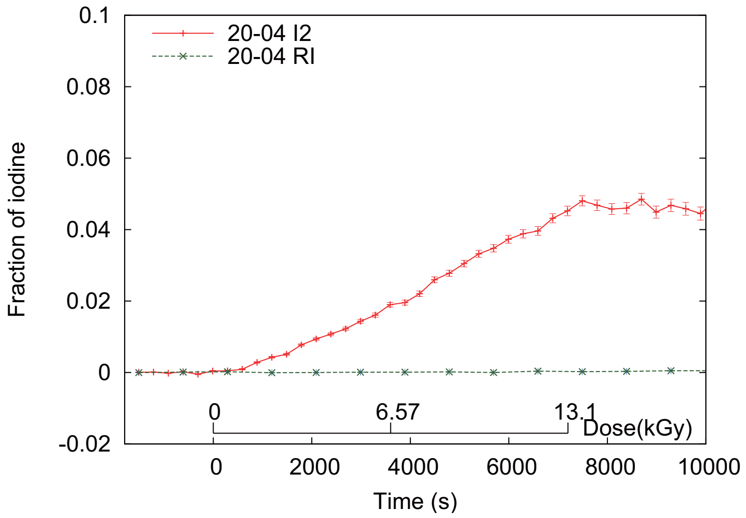


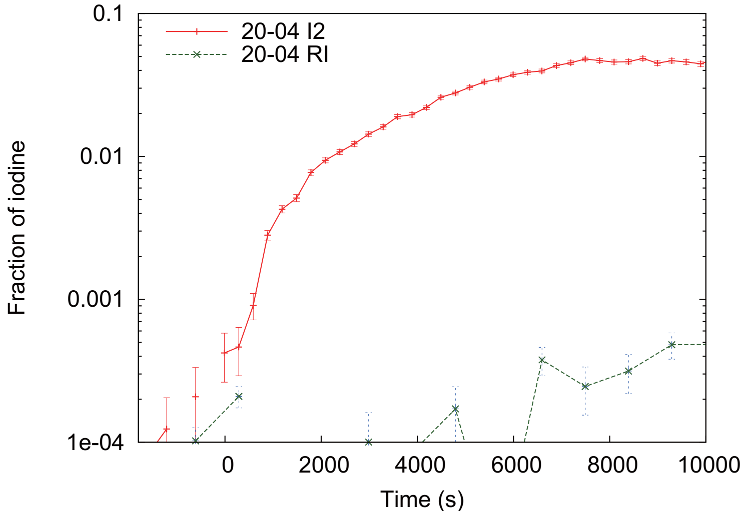
Distribution in filters



1-5: Ag filters
6-8: AgNO3 filters

Release history





Count efficiency(calibrated)= 0.1290 (Ag sampler)
0.1640 (AgNO3 sampler)

Tapered PTFE rings were inserted at the inlet of samplers.

Experimental condition

Run No.	**20-05**
Solution	pH=5.5 (Buffer: 0.1M B(OH)3-1.50e-5M NaOH), CsI 1e-4M, I-131 37kBq
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar
Temperature	40C
Gas	Air 1L/min
Dose rate	6.71 kGy/h
Date	2008/09/09

Irradiation schedule

Sweep start	10:50
Irrad. start	11:20
Irrad. end	13:20
Sweep end	13:50

Activity measurement

Run No.: 20-05 Date: 2008/09/09 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	08/09/09 11:10	1800.0	270490	3.0E-1	527.735	526.628	1.0000
Solution after irrad. 1mL	08/09/09 14:59	1800.0	111342	4.9E-1	217.234	219.778	4.030E-01
1: Ag-1	08/09/09 15:31	354.2	1041718	1.6E-1	27444.946	27799.599	5.357E-01
2: Ag-2	08/09/09 16:12	600.0	9089	2.1E+0	141.368	143.565	2.767E-03
3: Ag-3	08/09/09 16:23	600.0	777	1.6E+1	12.085	12.281	2.367E-04
4: Ag-4	08/09/09 16:35	163.4	47	1.2E+2	2.684	-	-
5: Ag-5	08/09/09 16:40	156.6	-64	-	0	-	-
6: AgNO3-1	08/09/09 15:39	1800.0	8022	3.0E+0	44.098	44.722	8.618E-04
7: AgNO3-2	08/09/09 16:43	163.5	-78	-	0	-	-
8: AgNO3-3	08/09/09 16:47	157.3	12	4.5E+2	0.755	-	-

Sum for filter 1-8	5.40E-01
Frac. in solution	4.03E-01
Loss	5.74E-02

* Fraction to total inventory

Activity values for solution samples were for whole solution

Sample mass(g)	Solution mass	Sample
Before	98.8	1.0027
After	94.6	0.9943

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
20-05	5.60	5.66	9.98E-05	3.94E-05	3.95E-01

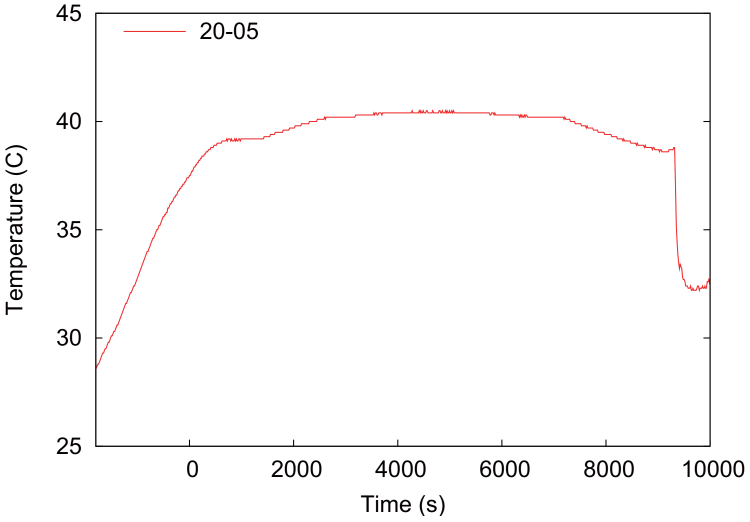
Filter fraction data

Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	5.357E-01	9.928E-01
2	2.767E-03	5.127E-03
3	2.367E-04	4.386E-04
4	-	-
5	-	-
AgNO3 (for Org-I)		
6	8.618E-04	1.597E-03
7	-	-
8	-	-

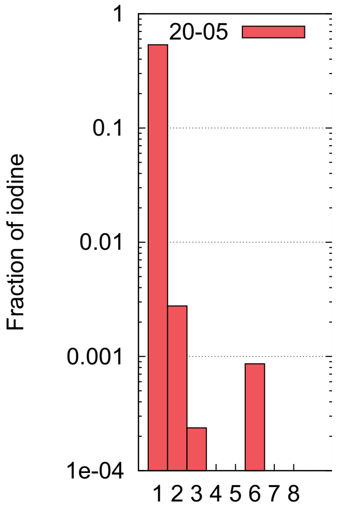
Adsorption was found at the up-stream coupler of the top sampler: 12cps(about 40Bq), PTFE ring: 2cps(about 10Bq)
Condensation in the top sampler.

Plots

Temperature	Initial	30 C
	During irrad.	39--40 C

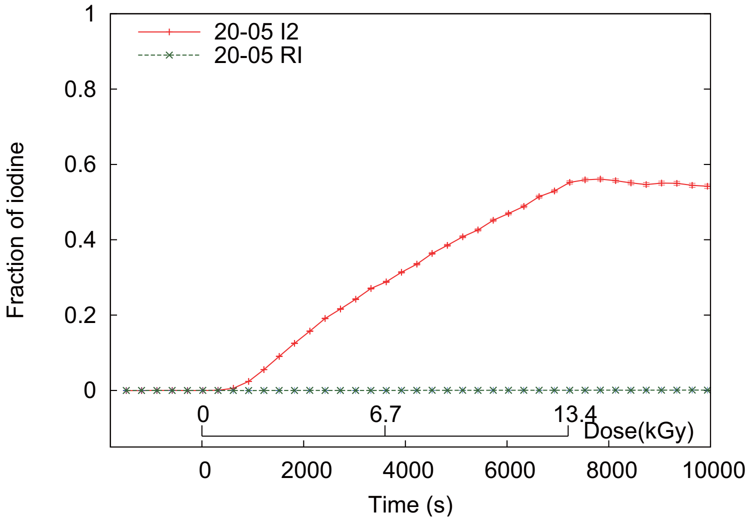


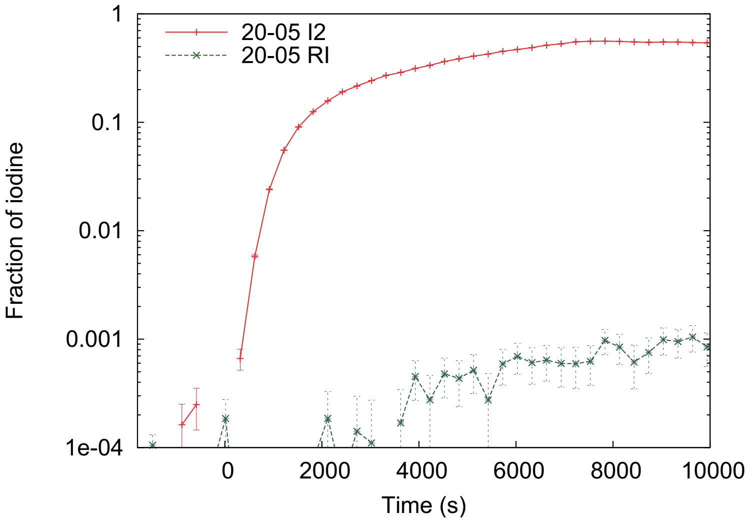
Distribution in filters



1-5: Ag filters
6-8: AgNO3 filters

Release history





Count efficiency(calibrated)= 0.1353 (Ag sampler)
0.1613 (AgNO3 sampler)

Tapered PTFE rings were inserted at the inlet of samplers.

Experimental condition

Run No.	**20-06**
Solution	pH=7.0 (Buffer: 0.1M B(OH)3-5.72e-4M NaOH), CsI 1e-4M, I-131 37kBq, MIBK 1e-3M
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar
Temperature	25C
Gas	N2-2%O2 1L/min
Dose rate	6.67 kGy/h
Date	2008/09/25

Irradiation schedule

Sweep start	11:03
Irrad. start	11:33
Irrad. end	13:33
Sweep end	14:03

Activity measurement

Run No.: 20-06 Date: 2008/09/25 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	08/09/25 11:08	1800.0	192871	3.6E-1	376.298	375.463	1.0000
Solution after irrad. 1mL	08/09/25 14:41	1800.0	176405	3.8E-1	344.176	347.830	9.343E-01
1: Ag-1	08/09/25 16:24	1201.1	141832	4.3E-1	1101.997	1120.268	2.991E-02
2: Ag-2	08/09/25 17:07	613.3	1403	9.6E+0	21.349	21.752	5.808E-04
3: Ag-3	08/09/25 17:19	631.6	1268	9.7E+0	18.735	19.103	5.101E-04
4: Ag-4	08/09/25 17:30	304.2	547	1.7E+1	16.783	17.121	4.571E-04
5: Ag-5	08/09/25 17:36	321.9	387	2.3E+1	11.220	11.451	3.057E-04
6: AgNO3-1	08/09/25 16:45	1221.5	69083	6.7E-1	559.622	569.623	1.521E-02
7: AgNO3-2	08/09/25 17:43	170.2	25	2.4E+2	1.453	-	-
8: AgNO3-3	08/09/25 17:47	153.1	-58	-	0	-	-

* Fraction to total inventory

Activity values for solution samples were for whole solution

Sum for filter 1-8	4.70E-02
Frac. in solution	9.34E-01
Loss	1.87E-02

Sample mass(g)	Solution mass	Sample
Before	99.6	0.9985
After	100.4	0.9980

*Before=before addition of MIBK solution

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
20-06	7.04	6.96	9.98E-05	7.70E-05	7.71E-01

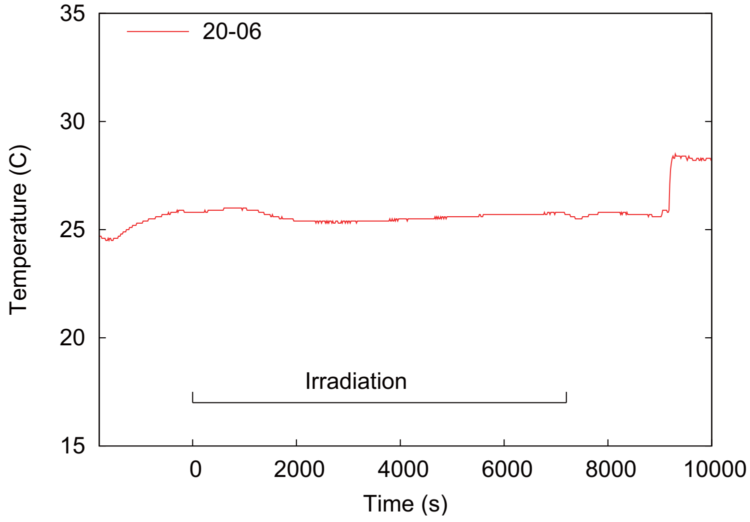
Filter fraction data

Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	2.991E-02	6.368E-01
2	5.808E-04	1.236E-02
3	5.101E-04	1.086E-02
4	4.571E-04	9.732E-03
5	3.057E-04	6.509E-03
AgNO3 (for Org-I)		
6	1.521E-02	3.238E-01
7	-	-
8	-	-

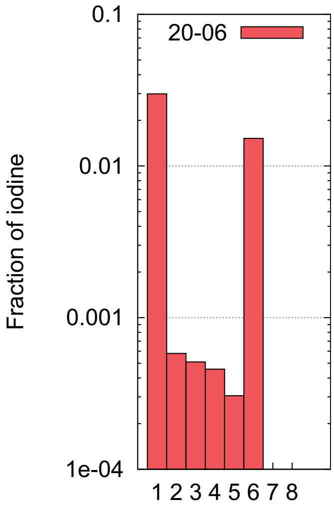
Adsorption was found at the up-stream coupler of the top sampler: 4cps(about 13Bq), PTFE ring: 1cps.

Plots

Temperature	Initial	25 C
	During irrad.	25--26 C

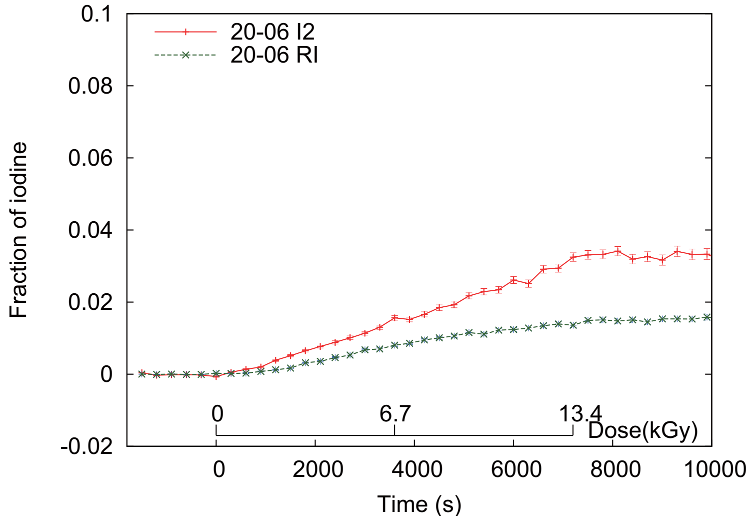


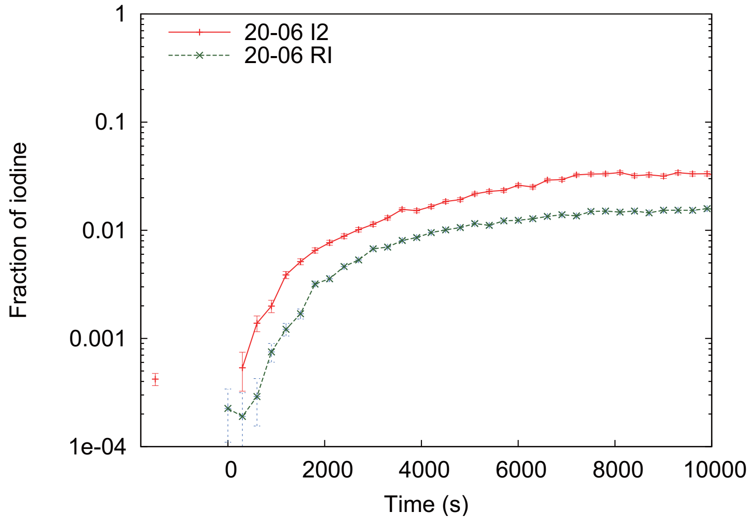
Distribution in filters



1-5: Ag filters
6-8: AgNO3 filters

Release history





Count efficiency(calibrated)=	0.1400 (Ag sampler)
	0.1795 (AgNO3 sampler)

Tapered PTFE rings were inserted at the inlet of samplers.

Experimental condition

Run No.	**20-07**
Solution	pH=7.0 (Buffer: 0.1M B(OH)3-5.72e-4M NaOH), CsI 1e-4M, I-131 37kBq
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar
Temperature	25C
Gas	N2-2%O2 1L/min
Dose rate	6.64 kGy/h
Date	2008/10/08

Irradiation schedule

Sweep start	10:02
Irrad. start	10:32
Irrad. end	12:32
Sweep end	13:02

Activity measurement

Run No.: 20-07 Date: 2008/10/08 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	08/10/08 10:22	1800.0	222115	3.4E-1	433.359	431.205	1.0000
Solution after irrad. 1mL	08/10/08 13:15	1800.0	189225	3.7E-1	369.188	371.188	8.578E-01
1: Ag-1	08/10/08 14:08	1311.5	702604	1.9E-1	4999.512	5041.369	1.192E-01
2: Ag-2	08/10/08 14:32	800.9	608	2.3E+1	7.085	7.152	1.692E-04
3: Ag-3	08/10/08 15:50	602.8	72	1.7E+2	1.115	-	-
4: Ag-4	08/10/08 15:24	638.2	319	3.6E+1	4.664	4.723	1.117E-04
5: Ag-5	08/10/08 15:36	300.9	-114	-	0	-	-
6: AgNO3-1	08/10/08 14:52	1800.0	2214	9.7E+0	12.171	12.308	2.911E-04
7: AgNO3-2	08/10/08 15:42	186.3	-22	-	0	-	-
8: AgNO3-3	08/10/08 15:46	174.0	21	2.8E+2	1.194	-	-

Sum for filter 1-8	1.20E-01
Frac. in solution	8.58E-01
Loss	2.24E-02

* Fraction to total inventory
Activity values for solution samples were for whole solution

Sample mass(g)	Solution mass	Sample
Before	98.7	1.0067
After	98.1	1.0041

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
20-07	7.03	7.02	9.98E-05	8.07E-05	8.08E-01

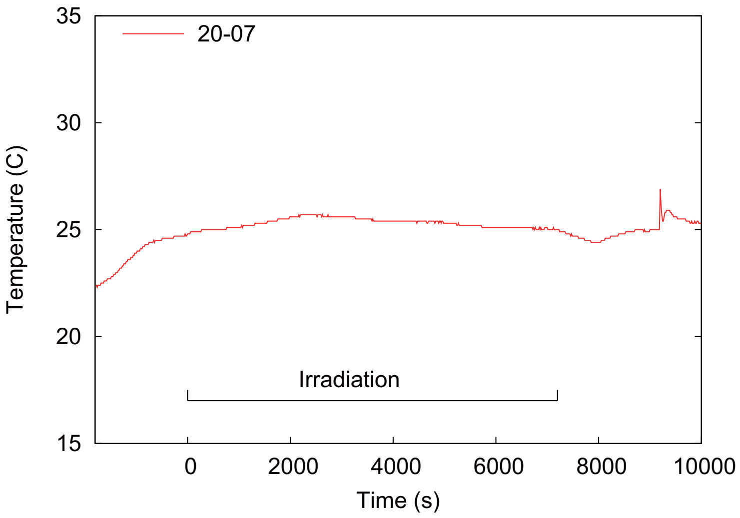
Filter fraction data

Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	1.192E-01	9.952E-01
2	1.692E-04	1.412E-03
3	-	-
4	1.117E-04	9.324E-04
5	-	-
AgNO3 (for Org-I)		
6	2.911E-04	2.430E-03
7	-	-
8	-	-

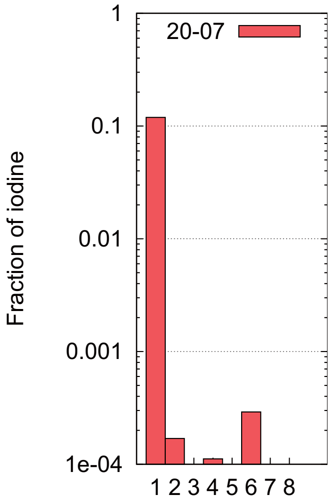
Adsorption was found at the up-stream coupler of the top sampler: 9cps(about 10Bq), PTFE ring: 1cps.
A small amount of Org-I release before irrad. Impurity of the tracer?

Plots

Temperature Initial 25 C
During irrad. 25--26 C

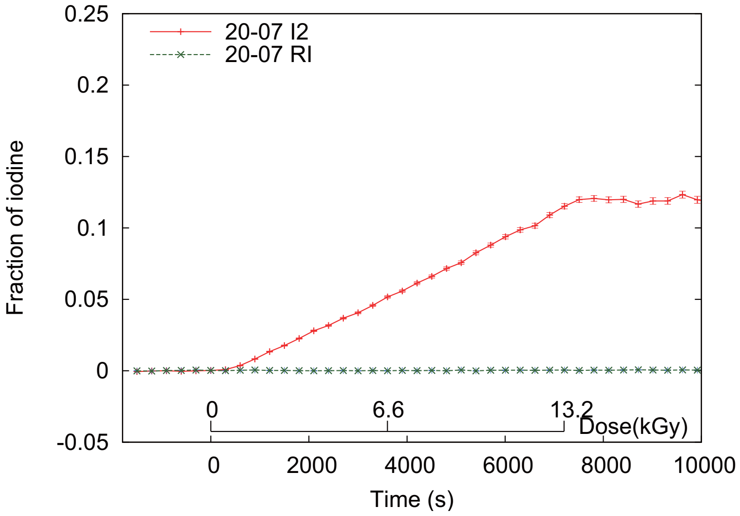


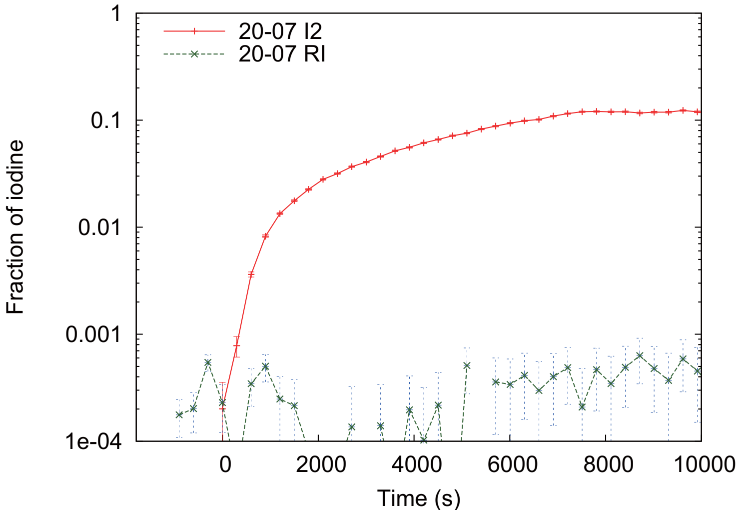
Distribution in filters



1-5: Ag filters
6-8: AgNO3 filters

Release history





Count efficiency(calibrated)= 0.1478 (Ag sampler)
0.2950 (AgNO3 sampler)

Tapered PTFE rings were inserted at the inlet of samplers.

Experimental condition

Run No.	**20-08**
Solution	pH=7.0 (Buffer: 0.1M B(OH)3-5.72e-4M NaOH), CsI 1e-4M, I-131 37kBq
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar
Temperature	25C
Gas	Air 1L/min
Dose rate	6.55 kGy/h
Date	2008/11/11

Irradiation schedule

Sweep start	10:38
Irrad. start	11:08
Irrad. end	13:08
Sweep end	13:38

Activity measurement

Run No.: 20-08 Date: 2008/11/11 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	08/11/11 10:26	1800.0	255500	3.1E-1	498.489	496.131	1.0000
Solution after irrad. 1mL	08/11/11 13:58	1800.0	232694	3.3E-1	453.999	457.637	9.049E-01
1: Ag-1	08/11/11 14:46	1387.9	558472	2.0E-1	3755.161	3795.390	7.637E-02
2: Ag-2	08/11/11 15:11	661.7	118	1.1E+2	1.664	-	-
3: Ag-3	08/11/11 15:55	301.2	126	5.9E+1	3.904	-	-
4: Ag-4	08/11/11 16:02	300.8	142	5.1E+1	4.405	-	-
5: Ag-5	08/11/11 16:08	223.9	11	5.9E+2	0.459	-	-
6: AgNO3-1	08/11/11 15:24	1800.0	1604	1.2E+1	8.817	8.934	1.798E-04
7: AgNO3-2	08/11/11 16:13	150.8	-86	-	0	-	-
8: AgNO3-3	08/11/11 16:16	162.5	98	5.0E+1	5.968	-	-

Sum for filter 1-8	7.66E-02
Frac. in solution	9.05E-01
Loss	1.85E-02

* Fraction to total inventory
Activity values for solution samples were for whole solution

Sample mass(g)	Solution mass	Sample
Before	100.9	1.0073
After	99.4	1.0115

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
20-08	6.97	6.91	9.98E-05	8.32E-05	8.33E-01

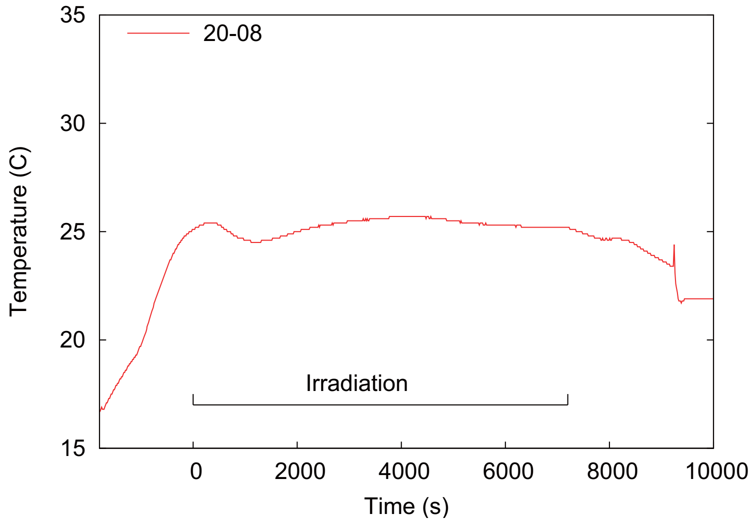
Filter fraction data

Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	7.637E-02	9.977E-01
2	-	-
3	-	-
4	-	-
5	-	-
AgNO3 (for Org-I)		
6	1.798E-04	2.348E-03
7	-	-
8	-	-

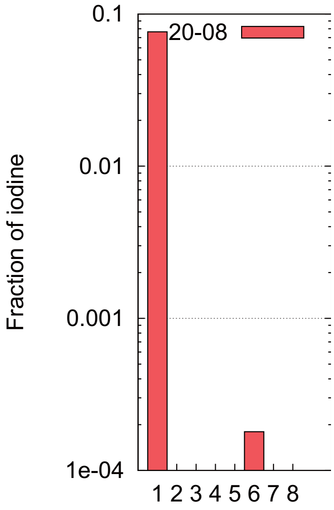
Adsorption was found at the up-stream coupler of the top sampler: 30cps(about 100Bq), PTFE ring:none.
The low room T (16C) affected T control. The final pH and I- sample T might be higher than the room T.

Plots

Temperature	Initial	17 C
	During irrad.	24-26 C

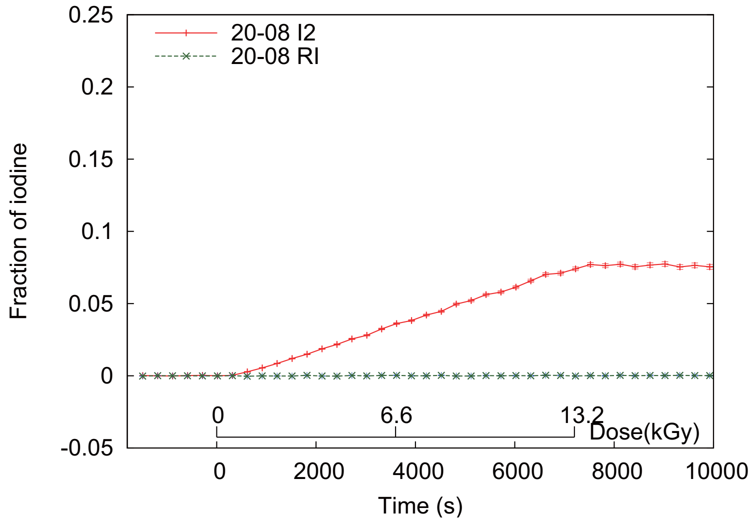


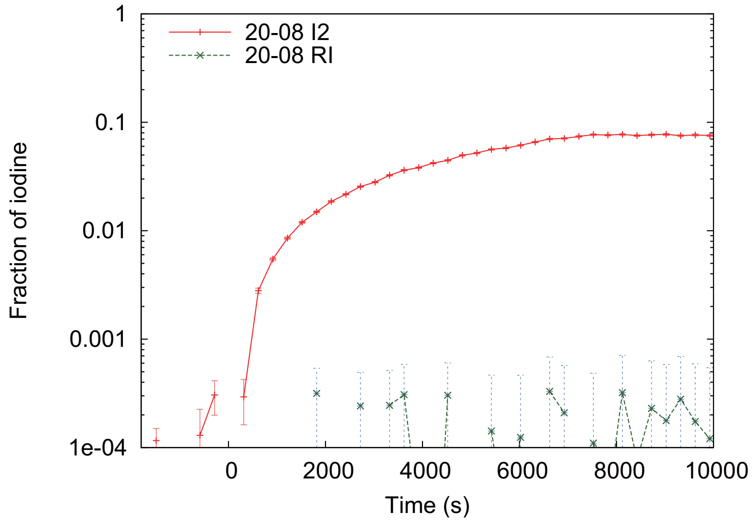
Distribution in filters



1-5: Ag filters
6-8: AgNO3 filters

Release history





Count efficiency(calibrated)= 0.1550 (Ag sampler)
0.1030 (AgNO3 sampler)

Tapered PTFE rings were inserted at the inlet of samplers.

Experimental condition

Run No.	**20-09**
Solution	pH=5.5 (Buffer: 0.1M B(OH)3-1.5e-5M NaOH), CsI 1e-4M, I-131 37kBq
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar
Temperature	25C
Gas	Air 1L/min
Dose rate	6.54 kGy/h
Date	2008/11/19

Irradiation schedule

Sweep start	10:22
Irrad. start	10:52
Irrad. end	12:52
Sweep end	13:22

Activity measurement

Run No.: 20-09 Date: 2008/11/19 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	08/11/19 10:16	1800.0	202046	3.6E-1	394.203	392.103	1.0000
Solution after irrad. 1mL	08/11/19 13:36	1800.0	44467	8.3E-1	86.757	87.336	2.161E-01
1: Ag-1	08/11/27 10:38	1800.0	2665093	9.0E-2	13817.363	27473.932	7.036E-01
2: Ag-2	08/11/19 14:31	1348.7	8706	2.6E+0	60.240	60.829	1.558E-03
3: Ag-3	08/11/19 15:27	601.2	315	3.8E+1	4.890	4.952	1.268E-04
4: Ag-4	08/11/19 15:38	201.9	2	3.4E+3	0.092	-	-
5: Ag-5	08/11/19 15:43	204.8	65	9.8E+1	2.962	-	-
6: AgNO3-1	08/11/19 14:55	1800.0	6765	3.5E+0	37.188	37.615	9.634E-04
7: AgNO3-2	08/11/19 15:48	201.1	54	1.1E+2	2.658	-	-
8: AgNO3-3	08/11/19 15:52	200.9	8	8.0E+2	0.394	-	-

Sum for filter 1-8	7.06E-01
Frac. in solution	2.16E-01
Loss	7.76E-02

* Fraction to total inventory

Activity values for solution samples were for whole solution

Sample mass(g)	Solution mass	Sample
Before	99.4	0.9982
After	97.8	1.0124

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
20-09	5.55	5.81	9.98E-05	1.87E-05	1.88E-01

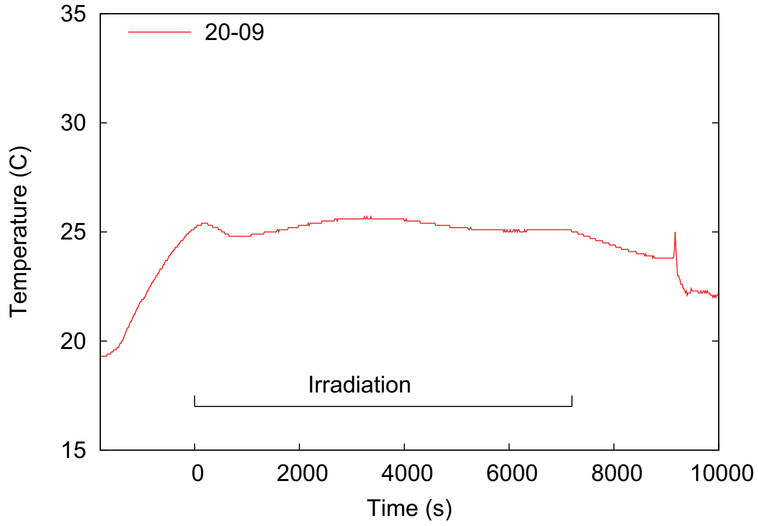
Filter fraction data

Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	7.036E-01	9.963E-01
2	1.558E-03	2.206E-03
3	1.268E-04	1.796E-04
4	-	-
5	-	-
AgNO3 (for Org-I)		
6	9.634E-04	1.364E-03
7	-	-
8	-	-

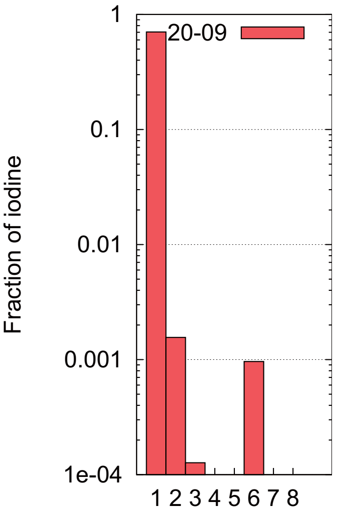
Ag-1 activity was too high and caused significant dead time. Re-measurement after 7 days were used.
Adsorption was found at the up-stream coupler of the top sampler: 45cps(about 160Bq), PTFE ring: 28cps (about 240Bq).
The low room T (16C) affected T control. The final pH and I-sample T might be higher than the room T.

Plots

Temperature	Initial	19 C
	During irrad.	25-26 C

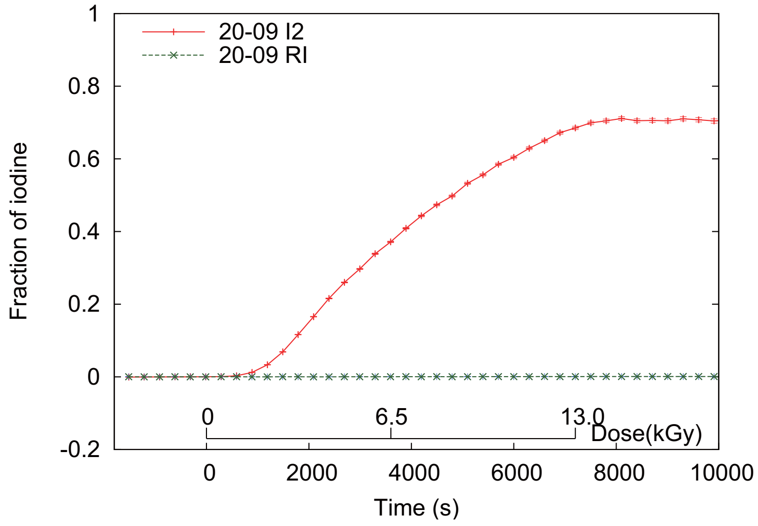


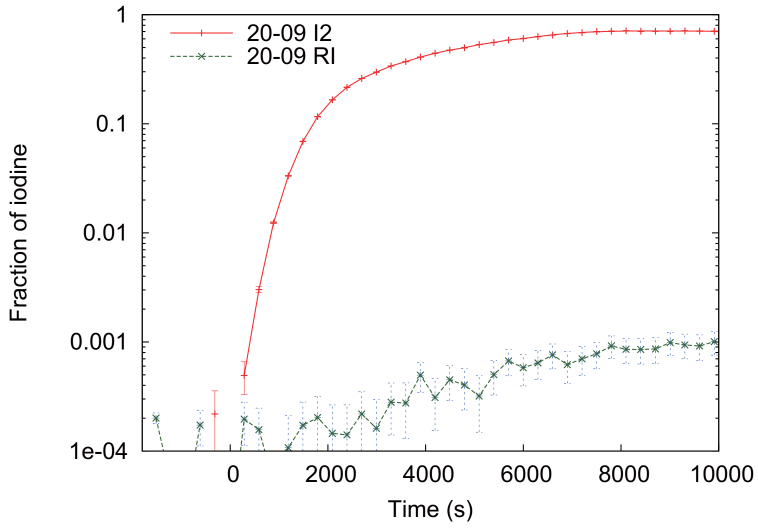
Distribution in filters



1-5: Ag filters
6-8: AgNO3 filters

Release history





Count efficiency(calibrated)=	0.1620 (Ag sampler)
	0.2550 (AgNO3 sampler)

Tapered PTFE rings were inserted at the inlet of samplers.

Experimental condition

Run No.	**20-10**
Solution	pH=7.0 (Buffer: 0.1M B(OH)3-5.72e-4M NaOH), CsI 1e-4M, I-131 37kBq, MIBK 1e-4M
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar
Temperature	25C
Gas	N2-2%O2 1L/min
Dose rate	6.51 kGy/h
Date	2008/11/28
Irradiation schedule	

Sweep start	11:31
Irrad. start	12:01
Irrad. end	14:01
Sweep end	14:31

Activity measurement

Run No.: 20-10 Date: 2008/11/28 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	08/11/28 11:24	1800.0	230399	3.3E-1	449.516	448.950	1.0000
Solution after irrad. 1mL	08/11/28 14:46	1800.0	205688	3.5E-1	401.309	405.691	8.895E-01
1: Ag-1	08/11/28 15:32	1800.0	943956	1.5E-1	4894.006	4961.132	1.099E-01
2: Ag-2	08/11/28 16:36	733.8	1036	1.3E+1	13.176	13.400	2.968E-04
3: Ag-3	08/11/28 16:50	153.8	-6	-	0	-	-
4: Ag-4	08/11/28 16:54	152.8	-9	-	0	-	-
5: Ag-5	08/11/28 16:58	159.2	-32	-	0	-	-
6: AgNO3-1	08/11/28 16:05	1800.0	13394	2.0E+0	73.629	74.787	1.657E-03
7: AgNO3-2	08/11/28 17:02	151.3	-86	-	0	-	-
8: AgNO3-3	08/11/28 17:05	168.1	27	2.1E+2	1.589	-	-

* Fraction to total inventory

Activity values for solution samples were for whole solution

Sum for filter 1-8	1.12E-01
Frac. in solution	8.90E-01
Loss	-1.39E-03

Sample mass(g)	Solution mass	Sample
Before	101.1	1.0054
After	99.7	1.0072

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
20-10	7.12	7.03	9.98E-05	8.68E-05	8.70E-01

Filter fraction data

Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	1.099E-01	9.825E-01
2	2.968E-04	2.654E-03
3	-	-
4	-	-
5	-	-
AgNO3 (for Org-I)		
6	1.657E-03	1.481E-02
7	-	-
8	-	-

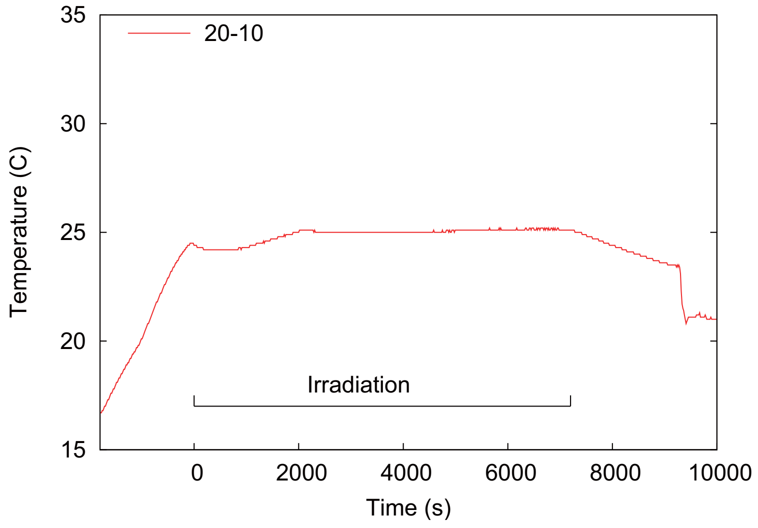
Adsorption was found at the up-stream coupler of the top sampler: 17cps(about 60Bq), PTFE ring: 3cps (about 20Bq).

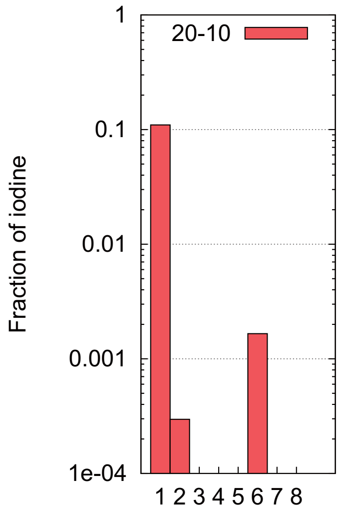
The low room T (14C) affected T control. The final pH and I- sample T might be higher than the room T.

Plots

Temperature	Initial	19 C
	During irrad.	24-26 C

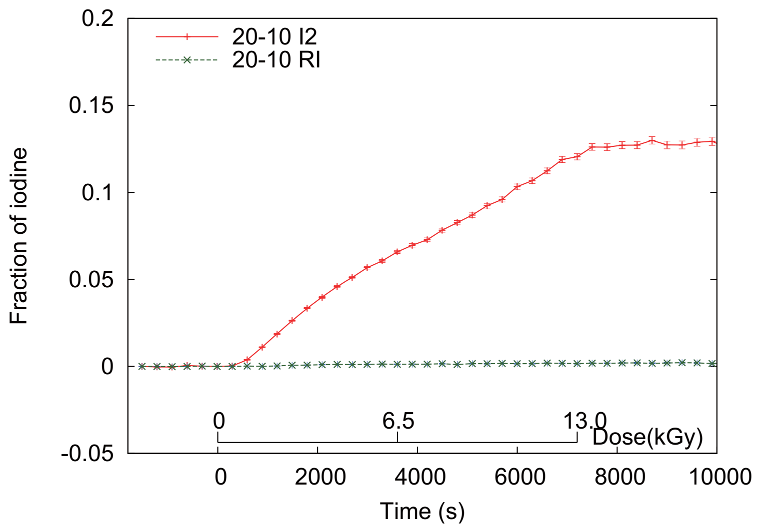
Distribution in filters

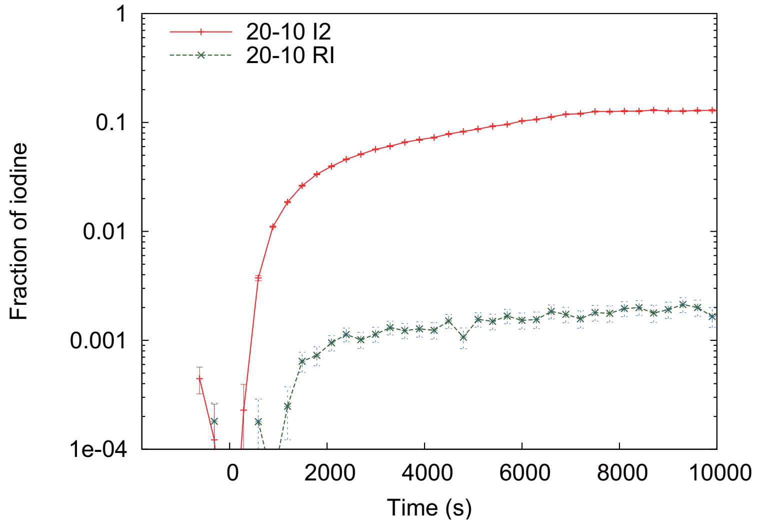




Release history

1-5: Ag filters
6-8: AgNO3 filters





Count efficiency(calibrated)= 0.1660 (Ag sampler)
0.2150 (AgNO3 sampler)

Tapered PTFE rings were inserted at the inlet of samplers.

Experimental condition

Run No.	**21-01**
Solution	pH=7.0 (Buffer: 0.1M B(OH)3-5.72e-4M NaOH), CsI 1e-4M, I-131 37kBq, NaCl 1e-2M
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar
Temperature	25C
Gas	Air 1L/min
Dose rate	6.09 kGy/h
Date	2009/06/02
Irradiation schedule	

Sweep start	10:45
Irrad. start	11:15
Irrad. end	13:15
Sweep end	13:45

Activity measurement

Run No.: 21-01 Date: 2009/06/02 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	09/06/02 10:54	1800.0	304286	2.8E-1	593.679	591.865	1.0000
Solution after irrad. 1mL	09/06/02 14:13	1800.0	277152	3.0E-1	540.739	545.563	9.123E-01
1: Ag-1	09/06/02 15:01	1410.5	791439	1.8E-1	5236.510	5297.435	9.069E-02
2: Ag-2	09/06/02 15:27	849.6	2531	6.0E+0	27.802	28.161	4.821E-04
3: Ag-3	09/06/02 16:16	860.4	1366	1.1E+1	14.817	15.053	2.577E-04
4: Ag-4	09/06/02 16:32	769.1	577	2.3E+1	7.002	7.120	1.219E-04
5: Ag-5	09/06/02 16:46	202.9	42	1.5E+2	1.931	-	-
6: AgNO3-1	09/06/02 15:43	1800.0	1647	1.2E+1	9.054	9.184	1.572E-04
7: AgNO3-2	09/06/02 16:51	154.5	-26	-	0	-	-
8: AgNO3-3	09/06/02 16:54	150.6	1	5.6E+3	0.066	-	-

Sum for filter 1-8	9.17E-02
Frac. in solution	9.12E-01
Loss	-3.98E-03

* Fraction to total inventory

Activity values for solution samples were for whole solution

Sample mass(g)	Solution mass	Sample
Before	98.9	1.0021
After	97.9	1.0023

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
21-01	7.06	-	9.98E-05	8.78E-05	8.80E-01

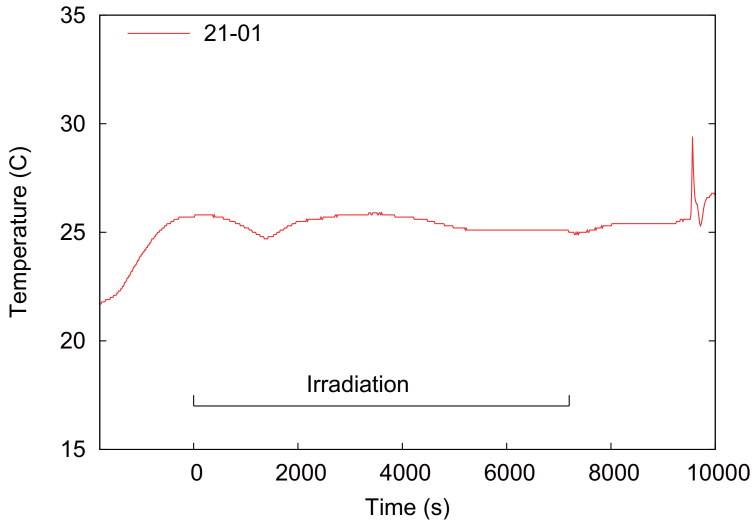
Filter fraction data

Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	9.069E-02	9.889E-01
2	4.821E-04	5.257E-03
3	2.577E-04	2.810E-03
4	1.219E-04	1.329E-03
5	-	-
AgNO3 (for Org-I)		
6	1.572E-04	1.714E-03
7	-	-
8	-	-

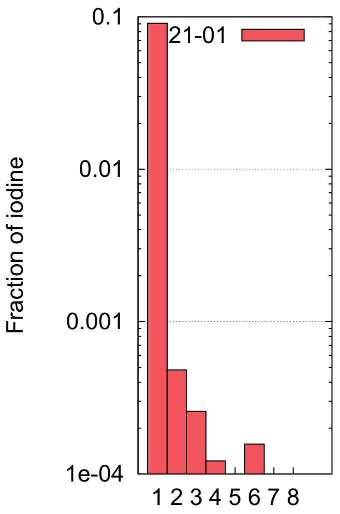
Adsorption was found at the up-stream coupler and PTFE ring of the top sampler: 12cps(about 40Bq).
pH measurement failed, the pH data was taken by the saved buffer solution (with Cl-) the day after. A pH drop of 0.2 was plausible during the exp. according to the measurement.

Plots

Temperature	Initial	17 C
	During irrad.	24-26 C

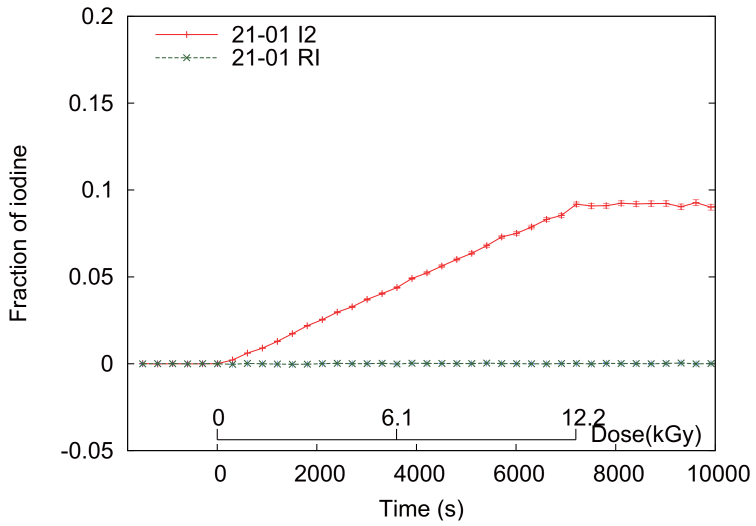


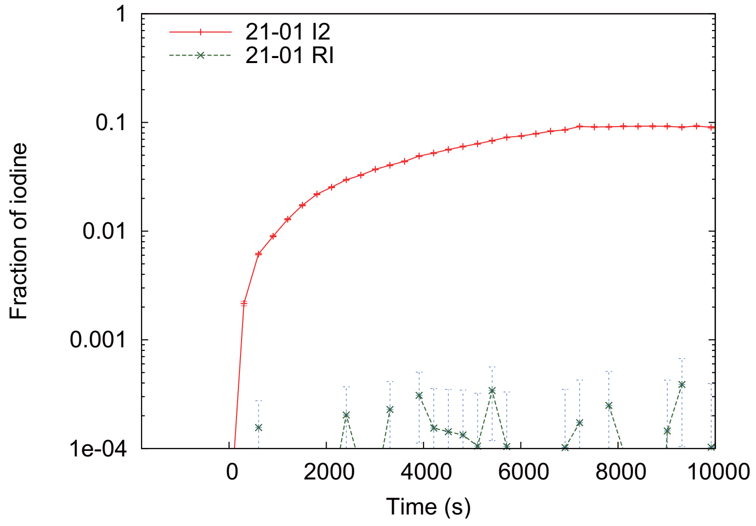
Distribution in filters



1-5: Ag filters
6-8: AgNO3 filters

Release history





Count efficiency(calibrated)= 0.1787 (Ag sampler)
0.1250 (AgNO3 sampler)

PTFE rings were inserted at the entrance of the samplers; The coupler in the up-stream of the top sampler was replaced with one made of PTFE (change of the position enhanced the efficiency)

Experimental condition

Run No.	**21-02**	
Solution	pH=7.0 (Buffer: 0.1M B(OH)3-5.72e-4M NaOH), CsI 1e-4M, I-131 37kBq, NaCl 1e-4M	
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar	
Temperature	25C	
Gas	Air 1L/min	
Dose rate	6.08 kGy/h	
Date	2009/06/10	
Irradiation schedule		
	Sweep start	11:11
	Irrad. start	11:41
	Irrad. end	13:41
	Sweep end	14:11

Activity measurement

Run No.: 21-02 Date: 2009/06/10 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	09/06/10 11:22	1800.0	216433	3.4E-1	422.273	421.690	1.0000
Solution after irrad. 1mL	09/06/10 14:24	1800.0	194422	3.6E-1	379.324	382.961	9.055E-01
1: Ag-1	09/06/10 15:17	1204.6	424896	2.4E-1	3291.791	3332.946	8.022E-02
2: Ag-2	09/06/10 16:13	602.0	458	2.5E+1	7.100	7.211	1.736E-04
3: Ag-3	09/06/10 16:24	300.9	29	2.8E+2	0.899	-	-
4: Ag-4	09/06/10 16:31	151.1	9	6.2E+2	0.556	-	-
5: Ag-5	09/06/10 16:35	159.4	-2	-	0	-	-
6: AgNO3-1	09/06/10 15:40	1800.0	1366	1.4E+1	7.509	7.616	1.833E-04
7: AgNO3-2	09/06/10 16:39	154.9	-9	-	0	-	-
8: AgNO3-3	09/06/10 16:43	153.9	-61	-	0	-	-

Sum for filter 1-8	8.06E-02
Frac. in solution	9.06E-01
Loss	1.39E-02

* Fraction to total inventory

Activity values for solution samples were for whole solution

Sample mass(g)	Solution mass	Sample
Before	99.2	1.0069
After	98.5	1.0027

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
21-02	6.97	6.93	9.98E-05	8.82E-05	8.84E-01

Filter fraction data

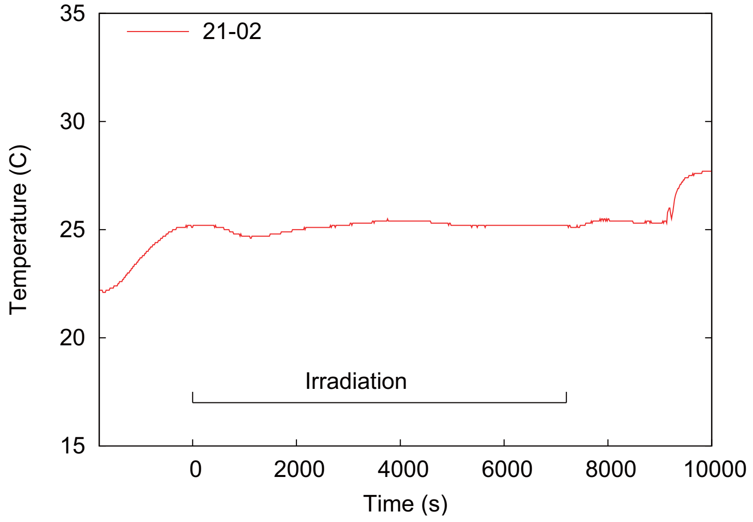
Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	8.022E-02	9.956E-01
2	1.736E-04	2.154E-03
3	-	-
4	-	-
5	-	-
AgNO3 (for Org-I)		
6	1.833E-04	2.275E-03
7	-	-
8	-	-

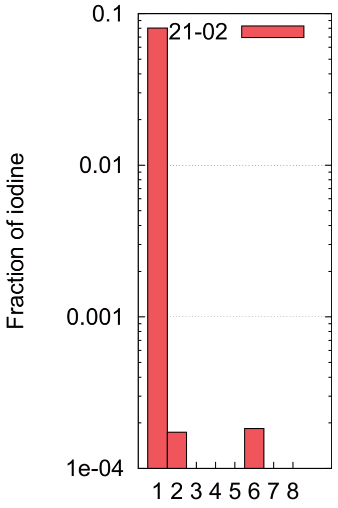
Adsorption was found at the up-stream coupler and PTFE ring of the top sampler: 6cps(about 20Bq).

Plots

Temperature	Initial	23 C
	During irrad.	24-26 C

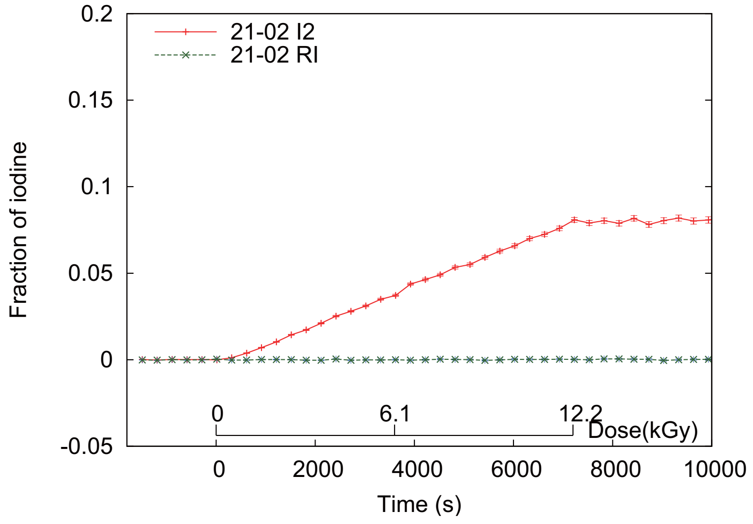
Distribution in filters

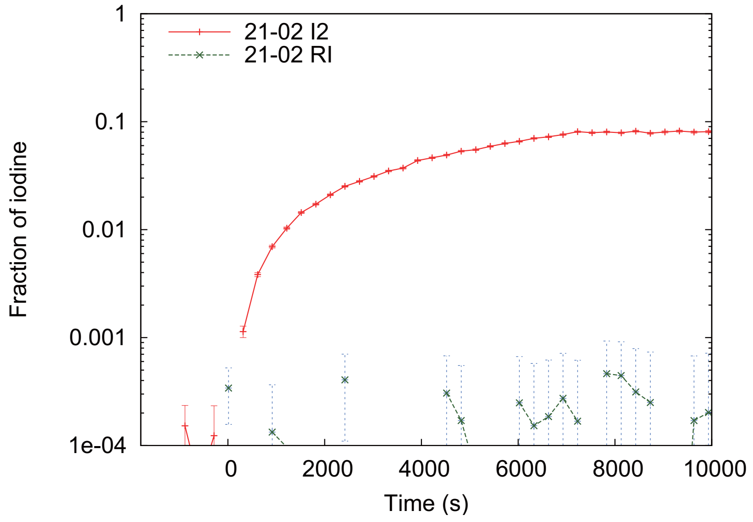




1-5: Ag filters
6-8: AgNO3 filters

Release history





Count efficiency(calibrated)=	0.1797 (Ag sampler) 0.1000 (AgNO3 sampler)

Experimental condition

Run No.	**21-03r**
Solution	pH=7.0 (Buffer: 0.1M B(OH)3-5.72e-4M NaOH), CsI 1e-4M, I-131 37kBq
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar
Temperature	25C
Gas	N2-5%H2 1L/min (4.80%H2 according to the analysis sheet)
Dose rate	5.82 kGy/h
Date	2009/10/08

Irradiation schedule

Sweep start	10:36
Irrad. start	11:06
Irrad. end	13:06
Sweep end	13:36

Activity measurement

Run No.: 21-03r Date: 2009/10/08 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	09/10/08 10:53	1800.0	218093	3.4E-1	425.511	424.186	1.0000
Solution after irrad. 1mL	09/10/08 13:48	1800.0	182974	3.7E-1	356.992	359.637	8.497E-01
1: Ag-1	09/10/08 14:45	1200.6	738337	1.8E-1	5739.071	5799.662	1.392E-01
2: Ag-2	09/10/08 15:07	725.1	271	4.2E+1	3.488	3.528	8.467E-05
3: Ag-3	09/10/08 15:55	241.3	11	5.5E+2	0.425	-	-
4: Ag-4	09/10/08 16:00	163.2	55	9.6E+1	3.146	-	-
5: Ag-5	09/10/08 16:05	154.8	-48	-	0	-	-
6: AgNO3-1	09/10/08 15:22	1800.0	1482	1.3E+1	8.147	8.254	1.981E-04
7: AgNO3-2	09/10/08 16:09	158.7	-88	-	0	-	-
8: AgNO3-3	09/10/08 16:13	204.6	-51	-	0	-	-

Sum for filter 1-8	1.39E-01
Frac. in solution	8.50E-01
Loss	1.09E-02

* Fraction to total inventory

Activity values for solution samples were for whole solution

Sample mass(g)	Solution mass	Sample
Before	99.2	1.0098
After	98.6	1.0015

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
21-03r	7.08	7.07	9.98E-05	7.73E-05	7.74E-01

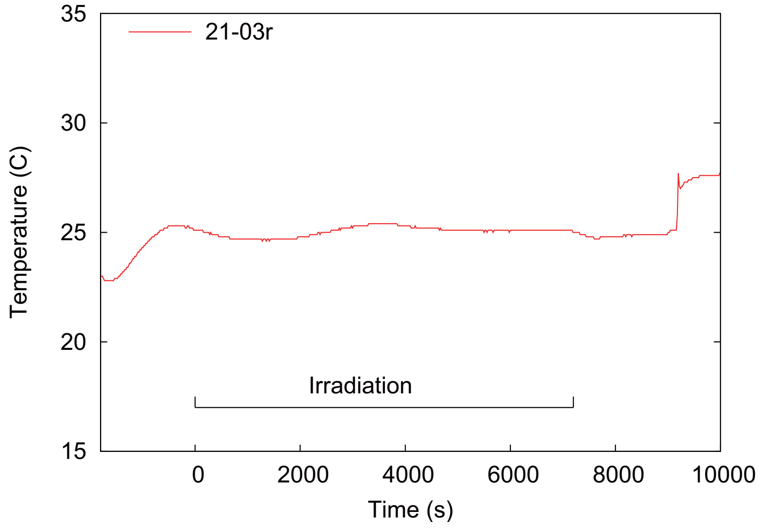
Filter fraction data

Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	1.392E-01	9.980E-01
2	8.467E-05	6.071E-04
3	-	-
4	-	-
5	-	-
AgNO3 (for Org-I)		
6	1.981E-04	1.420E-03
7	-	-
8	-	-

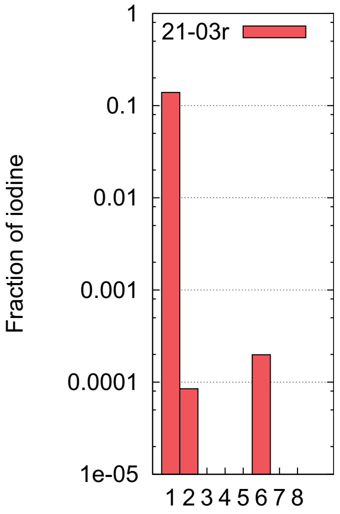
Adsorption was found at the up-stream coupler and PTFE ring of the top sampler: 6cps(about 20Bq).

Plots

Temperature	Initial	23 C
	During irrad.	24-26 C

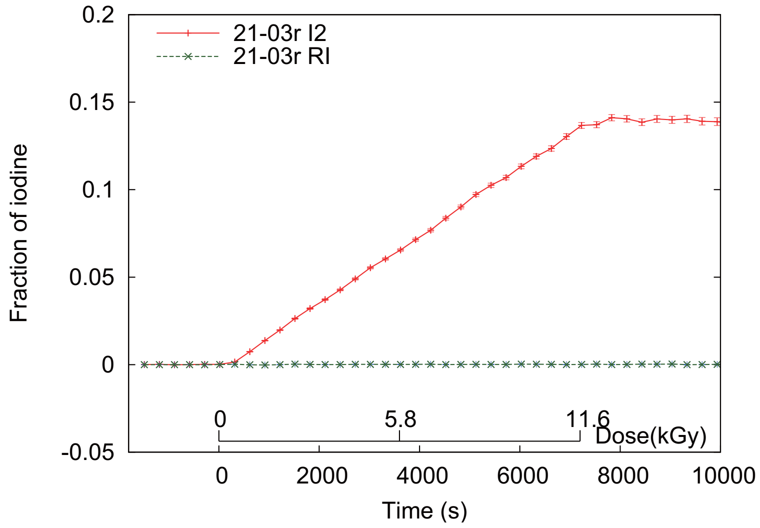


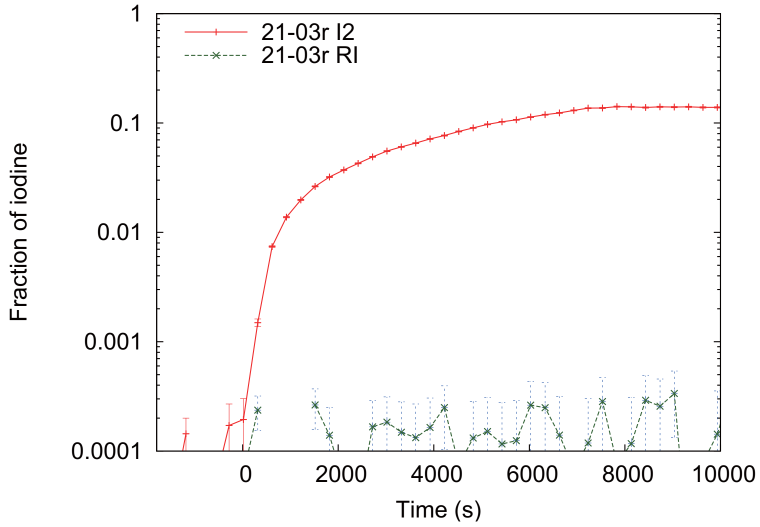
Distribution in filters



1-5: Ag filters
6-8: AgNO3 filters

Release history





Count efficiency(calibrated)= 0.2005 (Ag sampler)
0.2430 (AgNO3 sampler)

Experimental condition

Run No.	**21-04r**	
Solution	pH=7.0 (Buffer: 0.1M B(OH)3-5.72e-4M NaOH), CsI 1e-4M, I-131 37kBq	
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar	
Temperature	25C	
Gas	N2 1L/min	
Dose rate	5.78 kGy/h	
Date	2009/10/28	
Irradiation schedule		
	Sweep start	10:43
	Irrad. start	11:14
	Irrad. end	13:14
	Sweep end	13:44

Activity measurement

Run No.: 21-04r Date: 2009/10/28 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	09/10/28 10:33	1800.0	211976	3.4E-1	413.568	411.785	1.0000
Solution after irrad. 1mL	09/10/28 13:55	1800.0	157803	4.0E-1	307.879	310.290	7.501E-01
1: Ag-1	09/10/28 15:04	1093.9	1023802	1.5E-1	8734.210	8836.021	2.173E-01
2: Ag-2	09/10/28 15:25	1000.7	817	1.8E+1	7.619	7.717	1.898E-04
3: Ag-3	09/10/28 16:15	400.9	234	3.9E+1	5.447	5.532	1.361E-04
4: Ag-4	09/10/28 16:24	331.8	295	2.6E+1	8.297	8.431	2.074E-04
5: Ag-5	09/10/28 16:31	300.9	94	8.4E+1	2.916	-	-
6: AgNO3-1	09/10/28 15:43	1800.0	3506	5.8E+0	19.273	19.550	4.809E-04
7: AgNO3-2	09/10/28 16:38	600.6	276	3.5E+1	4.547	4.625	1.138E-04
8: AgNO3-3	09/10/28 16:50	151.8	-56	-	0	-	-

* Fraction to total inventory
Activity values for solution samples were for whole solution

Sum for filter 1-8	2.18E-01
Frac. in solution	7.50E-01
Loss	3.14E-02

Sample mass(g)	Solution mass	Sample
Before	99.9	1.0119
After	98.7	1.0043

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
21-04r	7.07	7.06	9.98E-05	7.35E-05	7.36E-01

Filter fraction data

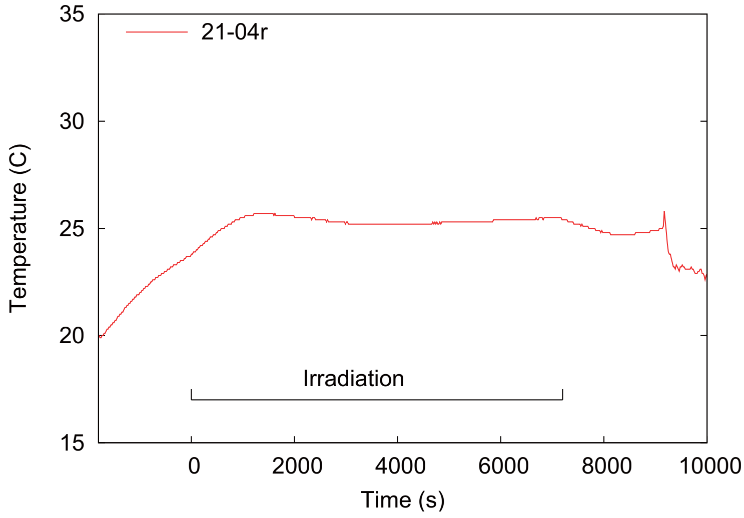
Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	2.173E-01	9.948E-01
2	1.898E-04	8.689E-04
3	1.361E-04	6.229E-04
4	2.074E-04	9.492E-04
5	-	-
AgNO3 (for Org-I)		
6	4.809E-04	2.201E-03
7	1.138E-04	5.207E-04
8	-	-

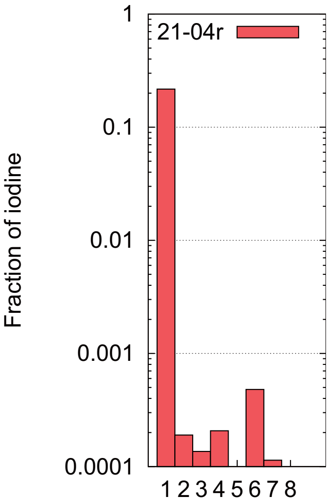
Adsorption was found at the up-stream coupler and PTFE ring of the top sampler: 2cps(about 8Bq).

Plots

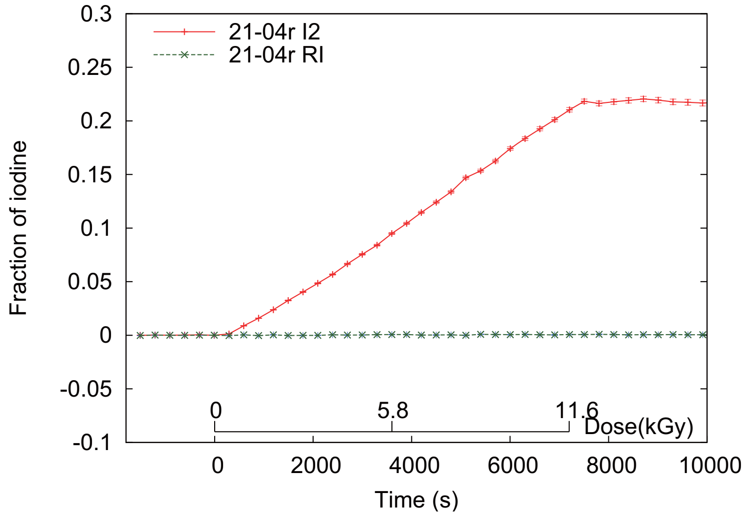
Temperature	Initial	20 C
	During irrad.	24-26 C

Distribution in filters

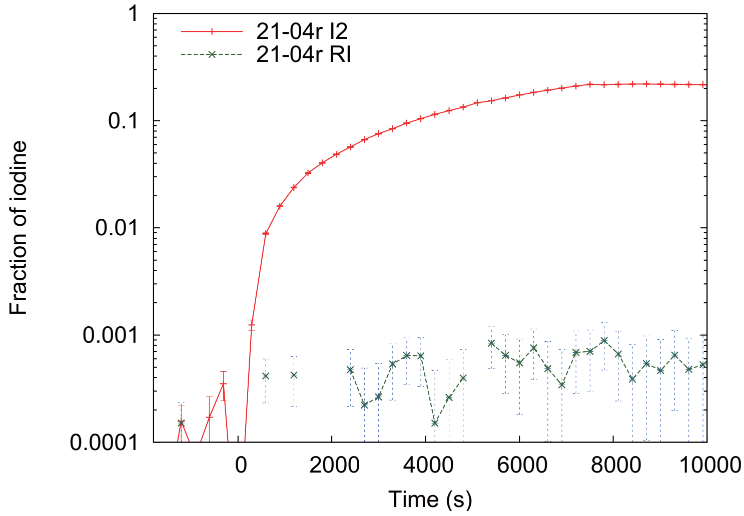




Release history



1-5: Ag filters
6-8: AgNO3 filters



Count efficiency(calibrated)= 0.1950 (Ag sampler)
0.1210 (AgNO3 sampler)

Experimental condition

Run No.	**22-01**
Solution	pH=7.0 (Buffer: 0.1M B(OH)3-5.72e-4M NaOH), CsI 1e-4M, I-131 37kBq
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar
Temperature	25C
Gas	Ar-5%H2 1L/min
Dose rate	5.29 kGy/h
Date	2010/06/29

Irradiation schedule

Sweep start	10:56
Irrad. start	11:27
Irrad. end	13:27
Sweep end	13:57

Activity measurement

Run No.: 22-01 Date: 2010/06/29 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	10/06/29 10:51	1800.0	256624	3.1E-1	500.682	499.062	1.0000
Solution after irrad. 1mL	10/06/29 14:10	1800.0	223128	3.3E-1	435.330	439.135	8.764E-01
1: Ag-1	10/06/29 15:00	1018.0	581645	2.1E-1	5331.961	5392.614	1.089E-01
2: Ag-2	10/06/29 15:51	1800.0	401	4.6E+1	2.079	2.110	4.262E-05
3: Ag-3	10/06/29 16:22	1348.8	287	5.5E+1	1.986	-	-
4: Ag-4	10/06/29 16:47	736.3	-38	-	0	-	-
5: Ag-5	10/06/29 17:00	470.9	-24	-	0	-	-
6: AgNO3-1	10/06/29 15:19	1800.0	1550	1.2E+1	8.520	8.631	1.743E-04
7: AgNO3-2	10/06/29 17:10	202.1	-24	-	0	-	-
8: AgNO3-3	10/06/29 17:14	201.7	74	8.0E+1	3.630	-	-

Sum for filter 1-8	1.091E-01
Frac. in solution	8.764E-01
Loss	1.446E-02

* Fraction to total inventory

Activity values for solution samples were for whole solution

Sample mass(g)	Solution mass	Sample
Before	99.5	1.0030
After	99.3	1.0050

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
22-01	7.01	7.03	9.98E-05	8.77E-05	8.79E-01

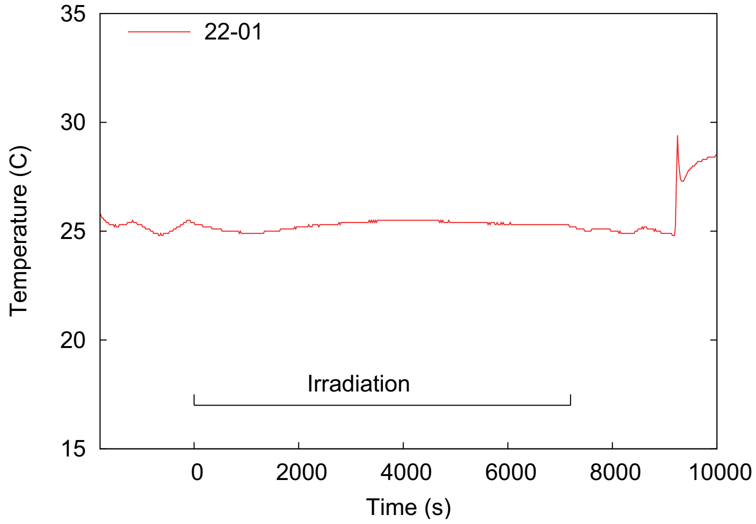
Filter fraction data

Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	1.089E-01	9.980E-01
2	4.262E-05	3.905E-04
3	-	-
4	-	-
5	-	-
AgNO3 (for Org-I)		
6	1.743E-04	1.597E-03
7	-	-
8	-	-

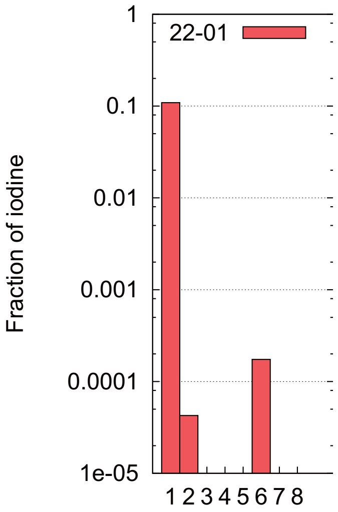
Adsorption was found at the up-stream coupler and PTFE ring of the top sampler: 4cps(about 15Bq).

Plots

Temperature	Initial	25 C
	During irrad.	25-25.5 C

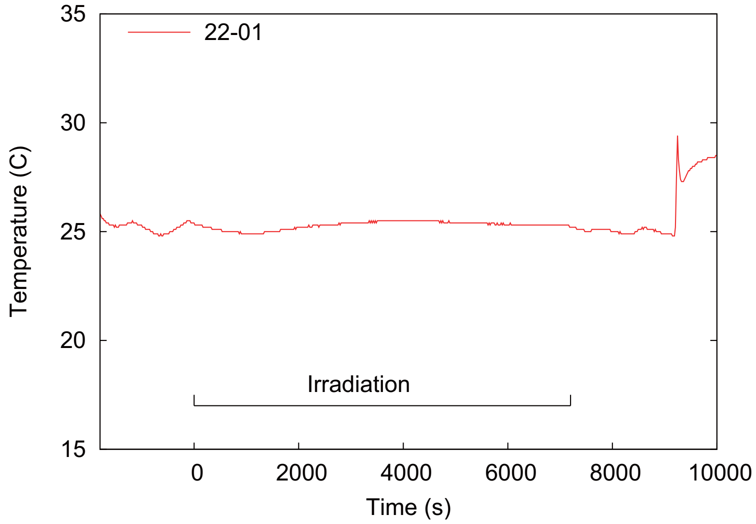


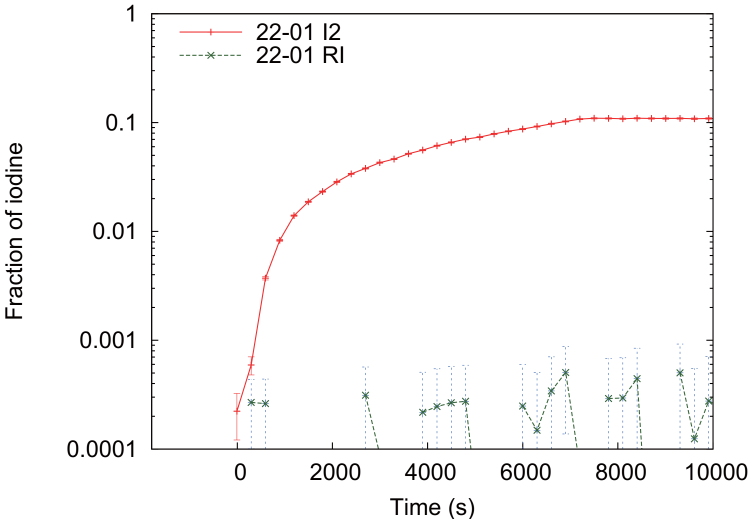
Distribution in filters



1-5: Ag filters
6-8: AgNO3 filters

Release history





Count efficiency(calibrated)= 0.1797 (Ag sampler)
0.0970 (AgNO3 sampler)

Experimental condition

Run No.	**22-02**	
Solution	pH=7.0 (Buffer: 0.1M B(OH)3-5.72e-4M NaOH), CsI 1e-4M, I-131 37kBq	
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar	
Temperature	25C	
Gas	Ar 1L/min	
Dose rate	5.29 kGy/h	
Date	2010/07/01	
Irradiation schedule		
	Sweep start	10:56
	Irrad. start	11:27
	Irrad. end	13:27
	Sweep end	13:57

Activity measurement

Run No.: 22-02 Date: 2010/07/01 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	10/07/01 10:52	1800.0	207612	3.5E-1	405.062	403.776	1.0000
Solution after irrad. 1mL	10/07/01 14:07	1800.0	163853	3.9E-1	319.686	322.422	7.941E-01
1: Ag-1	10/07/01 15:00	1079.9	803478	1.8E-1	6943.585	7022.789	1.753E-01
2: Ag-2	10/07/01 15:20	627.2	304	3.6E+1	4.523	4.579	1.143E-04
3: Ag-3	10/07/01 16:03	319.3	67	1.1E+2	1.958	-	-
4: Ag-4	10/07/01 16:09	307.5	113	6.4E+1	3.429	-	-
5: Ag-5	10/07/01 16:16	315.3	38	2.0E+2	1.125	-	-
6: AgNO3-1	10/07/01 15:31	1800.0	1927	1.1E+1	10.593	10.738	2.681E-04
7: AgNO3-2	10/07/01 16:23	300.8	-143	-	0	-	-
8: AgNO3-3	10/07/01 16:28	300.7	-34	-	0	-	-

* Fraction to total inventory

Activity values for solution samples were for whole solution

Sum for filter 1-8	1.757E-01
Frac. in solution	7.941E-01
Loss	3.014E-02

Sample mass(g)	Solution mass	Sample
Before	99.1	0.9991
After	98.9	1.0026

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
22-02	6.98	7.04	9.98E-05	7.36E-05	7.37E-01

Filter fraction data

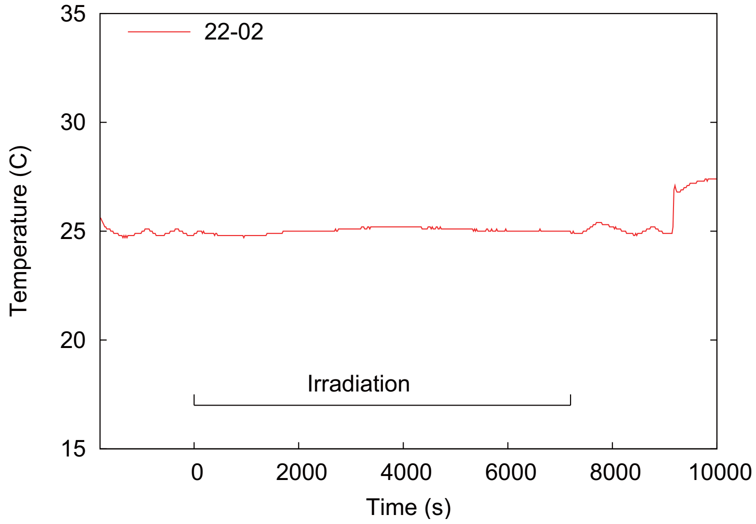
Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	1.753E-01	9.978E-01
2	1.143E-04	6.506E-04
3	-	-
4	-	-
5	-	-
AgNO3 (for Org-I)		
6	2.681E-04	1.526E-03
7	-	-
8	-	-

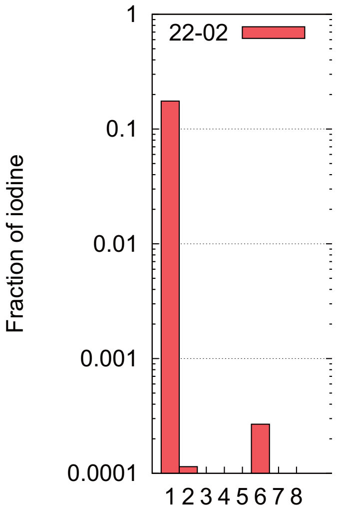
Adsorption was found at the up-stream coupler and PTFE ring of the top sampler: 100cps(about 360Bq).

Plots

Temperature	Initial	25 C
	During irrad.	25-25.5 C

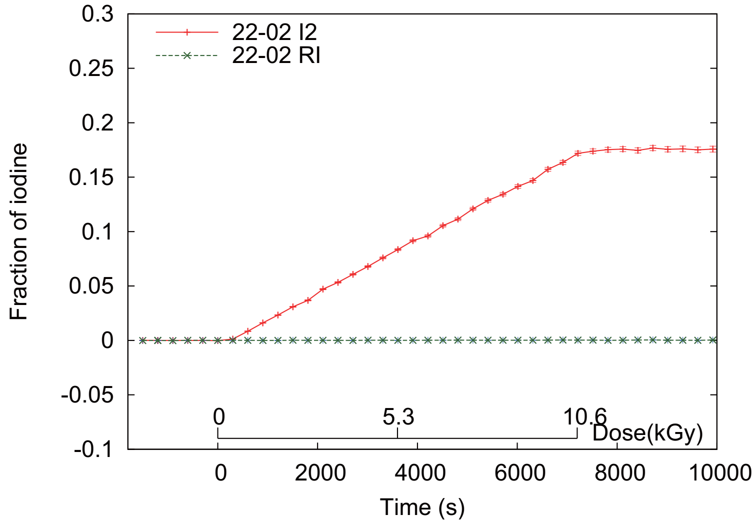
Distribution in filters

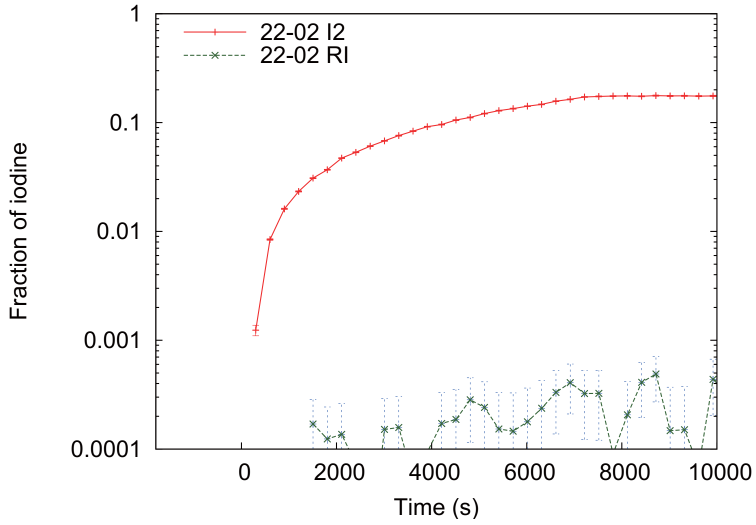




1-5: Ag filters
6-8: AgNO3 filters

Release history





Count efficiency(calibrated)= 0.1585 (Ag sampler)
0.2000 (AgNO3 sampler)

Experimental condition

Run No.	**22-03**	
Solution	pH=7.0 (Buffer: 0.1M B(OH)3-5.72e-4M NaOH), CsI 1e-4M, I-131 37kBq	
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar	
Temperature	25C	
Gas	Ar-3%H2 1L/min	
Dose rate	5.27 kGy/h	
Date	2010/07/08	
Irradiation schedule		
	Sweep start	11:06
	Irrad. start	11:36
	Irrad. end	13:36
	Sweep end	14:06

Activity measurement

Run No.: 22-03 Date: 2010/07/08 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	10/07/08 10:59	1800.0	249619	3.1E-1	487.015	485.672	1.0000
Solution after irrad. 1mL	10/07/08 14:17	1800.0	207120	3.5E-1	404.094	407.796	8.425E-01
1: Ag-1	10/07/08 15:04	1224.3	832348	1.7E-1	6344.365	6418.738	1.345E-01
2: Ag-2	10/07/08 15:26	918.0	514	2.4E+1	5.225	5.293	1.109E-04
3: Ag-3	10/07/08 16:17	272.8	64	1.1E+2	2.190	-	-
4: Ag-4	10/07/08 16:23	154.6	-16	-	0	-	-
5: Ag-5	10/07/08 16:27	203.7	15	3.9E+2	0.687	-	-
6: AgNO3-1	10/07/08 15:42	1800.0	2300	8.4E+0	12.643	12.824	2.688E-04
7: AgNO3-2	10/07/08 16:32	200.7	-90	-	0	-	-
8: AgNO3-3	10/07/08 16:37	223.8	85	6.7E+1	3.758	-	-

Sum for filter 1-8	1.349E-01
Frac. in solution	8.425E-01
Loss	2.259E-02

* Fraction to total inventory

Activity values for solution samples were for whole solution

Sample mass(g)	Solution mass	Sample
Before	98.8	1.0058
After	98.7	1.0014

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
22-03	6.99	7.01	9.98E-05	9.02E-05	9.04E-01

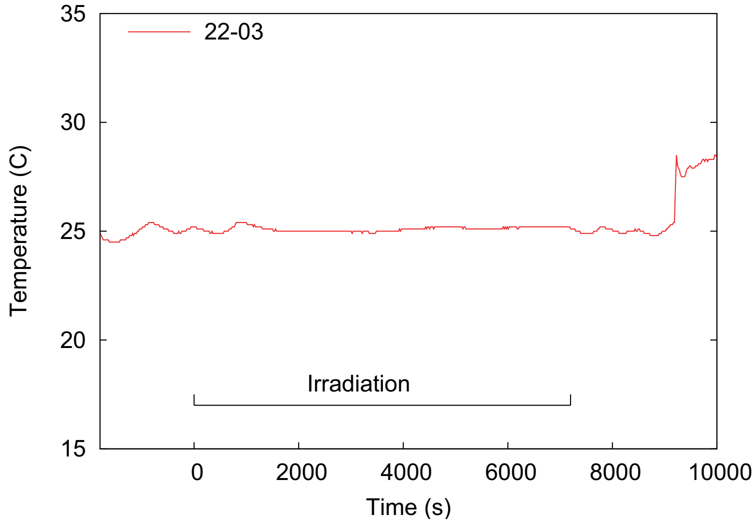
Filter fraction data

Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	1.345E-01	9.972E-01
2	1.109E-04	8.223E-04
3	-	-
4	-	-
5	-	-
AgNO3 (for Org-I)		
6	2.688E-04	1.992E-03
7	-	-
8	-	-

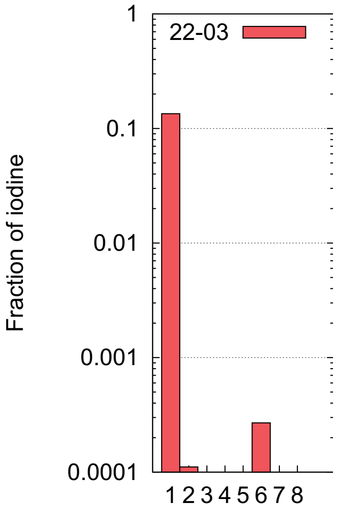
Adsorption was found at the up-stream coupler and PTFE ring of the top sampler: 1cps(about 4Bq).

Plots

Temperature	Initial	25 C
	During irrad.	25-25.5 C

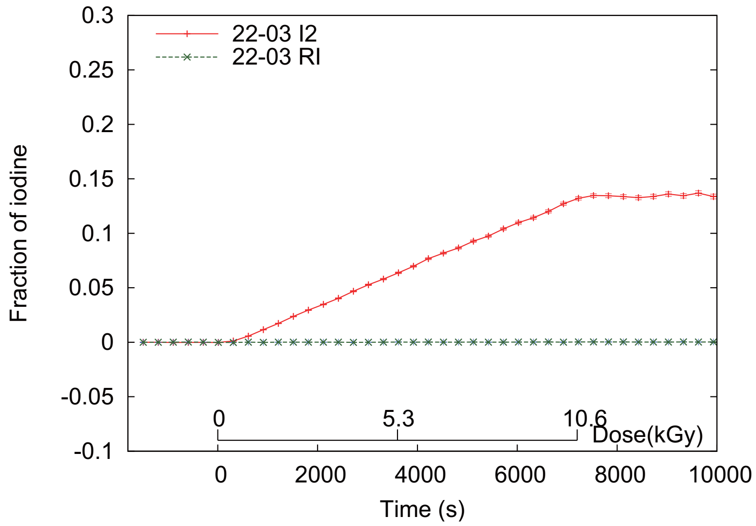


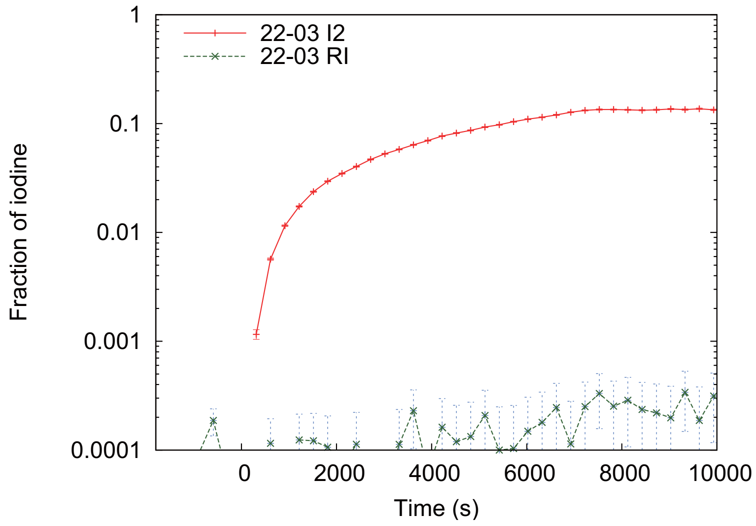
Distribution in filters



1-5: Ag filters
6-8: AgNO3 filters

Release history





Count efficiency(calibrated)= 0.1833 (Ag sampler)
0.2310 (AgNO3 sampler)

Experimental condition

Run No.	**22-04**
Solution	pH=7.0 (Buffer: 0.1M B(OH)3-5.72e-4M NaOH), CsI 1e-4M, MIBK 1e-4M (Irrad. start直前に0.05M MIBK 0.2mL添加), I-131 37kBq
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar
Temperature	25C
Gas	N2-5%O2 1L/min
Dose rate	5.26 kGy/h
Date	2010/07/13

Irradiation schedule

Sweep start	11:33
Irrad. start	12:03
Irrad. end	14:03
Sweep end	14:33

Activity measurement

Run No.: 22-04 Date: 2010/07/13 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	10/07/13 12:07	1800.0	212280	3.4E-1	414.170	414.717	1.0000
Solution after irrad. 1mL	10/07/13 14:43	1800.0	187032	3.6E-1	364.906	368.825	8.879E-01
1: Ag-1	10/07/13 15:26	1422.4	571509	2.1E-1	3749.615	3798.958	9.276E-02
2: Ag-2	10/07/13 15:52	600.6	541	1.9E+1	8.406	8.526	2.082E-04
3: Ag-3	10/07/13 16:44	180.5	-20	-	0	-	-
4: Ag-4	10/07/13 16:48	204.6	13	4.9E+2	0.593	-	-
5: Ag-5	10/07/13 16:52	181.2	35	1.6E+2	1.803	-	-
6: AgNO3-1	10/07/13 16:03	1800.0	5238	4.1E+0	28.794	29.243	7.140E-04
7: AgNO3-2	10/07/13 16:34	181.1	-40	-	0	-	-
8: AgNO3-3	10/07/13 16:38	284.0	42	1.6E+2	1.464	-	-

Sum for filter 1-8	9.368E-02
Frac. in solution	8.879E-01
Loss	1.841E-02

* Fraction to total inventory
Activity values for solution samples were for whole solution

Sample mass(g)	Solution mass	Sample
Before	99.3	1.0055
After	99.2	1.0061

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
22-04	6.99	6.97	9.98E-05	7.94E-05	7.96E-01

Filter fraction data

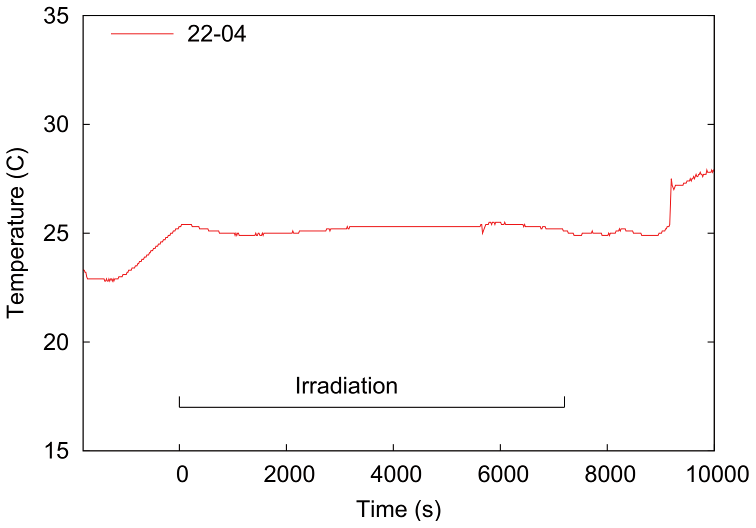
Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	9.276E-02	9.902E-01
2	2.082E-04	2.222E-03
3	-	-
4	-	-
5	-	-
AgNO3 (for Org-I)		
6	7.140E-04	7.622E-03
7	-	-
8	-	-

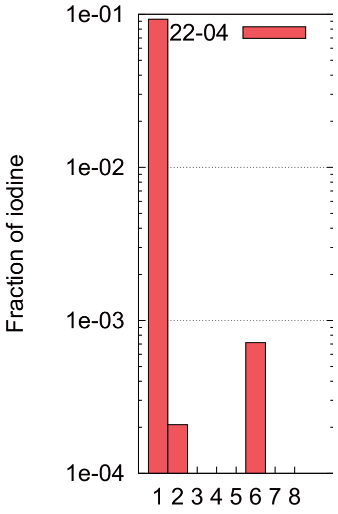
Adsorption was found at the up-stream coupler and PTFE ring of the top sampler: 2cps(about 8Bq).

Plots

Temperature Initial 23 C
During irrad. 25-25.5 C

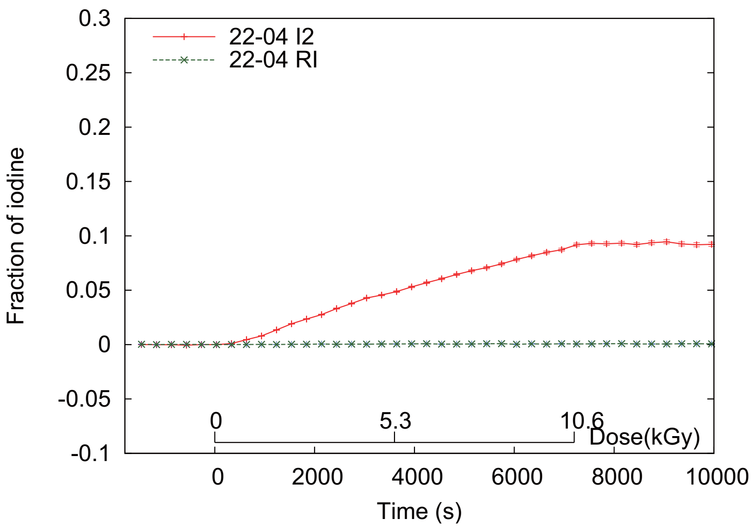
Distribution in filters

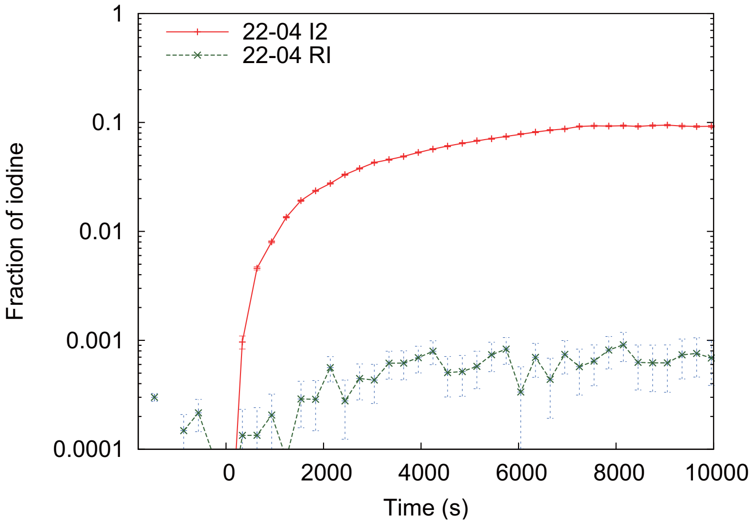




1-5: Ag filters
6-8: AgNO3 filters

Release history





Count efficiency(calibrated)= 0.1912 (Ag sampler)
0.1910 (AgNO3 sampler)

Experimental condition

Run No.	**22-05**
Solution	pH=7.0 (Buffer: 0.1M B(OH)3-5.72e-4M NaOH), CsI 1e-4M, I-131 37kBq
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar
Temperature	25C
Gas	N2-5%O2 1L/min
Dose rate	5.26 kGy/h
Date	2010/07/16

Irradiation schedule

Sweep start	11:31
Irrad. start	12:01
Irrad. end	14:01
Sweep end	14:31

Activity measurement

Run No.: 22-05 Date: 2010/07/16 (Activity compensation at 12:00)

	Start of measurement	Period (s)	Net count	Relative error(%)	Activity (Bq)	Compensated activity(Bq)	Iodine fraction*
Solution before irrad. 1mL	10/07/16 11:26	1800.0	205058	3.5E-1	400.075	399.619	1.0000
Solution after irrad. 1mL	10/07/16 14:43	1800.0	186046	3.7E-1	362.986	366.884	8.998E-01
1: Ag-1	10/07/16 15:29	1546.3	568186	2.1E-1	3429.159	3475.126	8.733E-02
2: Ag-2	10/07/16 16:32	900.3	511	2.4E+1	5.297	5.386	1.354E-04
3: Ag-3	10/07/16 16:48	162.1	31	1.8E+2	1.785	-	-
4: Ag-4	10/07/16 16:52	160.6	76	6.4E+1	4.417	-	-
5: Ag-5	10/07/16 17:00	160.5	42	1.3E+2	2.441	-	-
6: AgNO3-1	10/07/16 15:59	1800.0	1281	1.4E+1	7.042	7.150	1.797E-04
7: AgNO3-2	10/07/16 17:04	161.0	23	2.3E+2	1.414	-	-
8: AgNO3-3	10/07/16 17:08	160.7	45	1.2E+2	2.771	-	-

Sum for filter 1-8	8.764E-02
Frac. in solution	8.998E-01
Loss	1.252E-02

* Fraction to total inventory

Activity values for solution samples were for whole solution

Sample mass(g)	Solution mass	Sample
Before	99.9	1.0032
After	99.3	1.0174

Chemical measurements

Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
22-05	7.01	7.02	9.98E-05	7.95E-05	7.97E-01

Filter fraction data

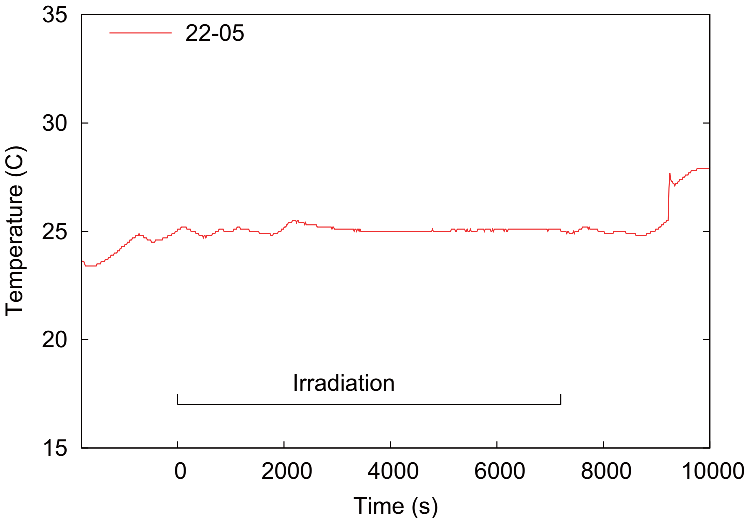
Filter No.	Frac. in whole	Frac. in filters
Ag (for I2)		
1	8.733E-02	9.964E-01
2	1.354E-04	1.544E-03
3	-	-
4	-	-
5	-	-
AgNO3 (for Org-I)		
6	1.797E-04	2.050E-03
7	-	-
8	-	-

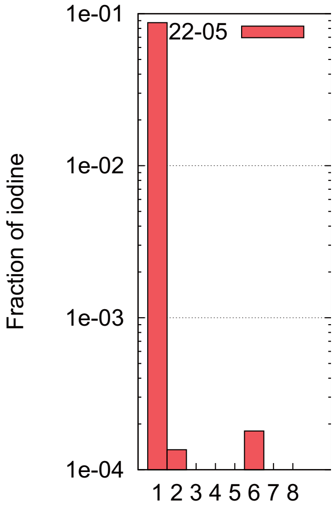
Adsorption was found at the up-stream coupler and PTFE ring of the top sampler: 1.5cps(about 5Bq).

Plots

Temperature	Initial	23 C
	During irrad.	24.5-25.5 C

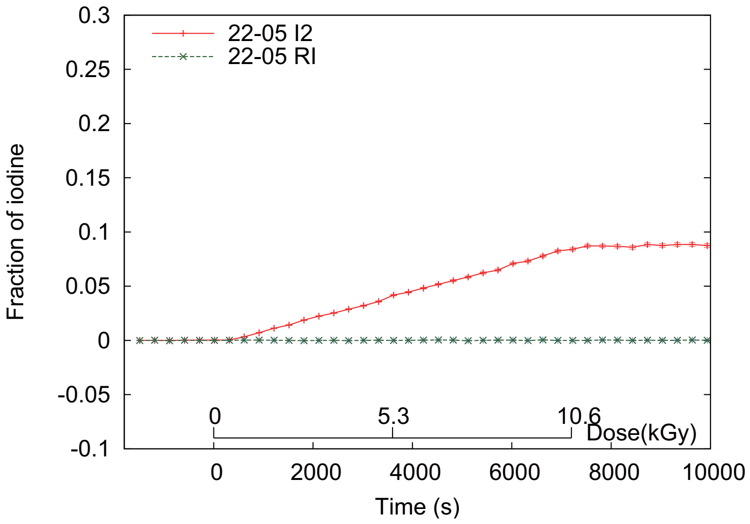
Distribution in filters

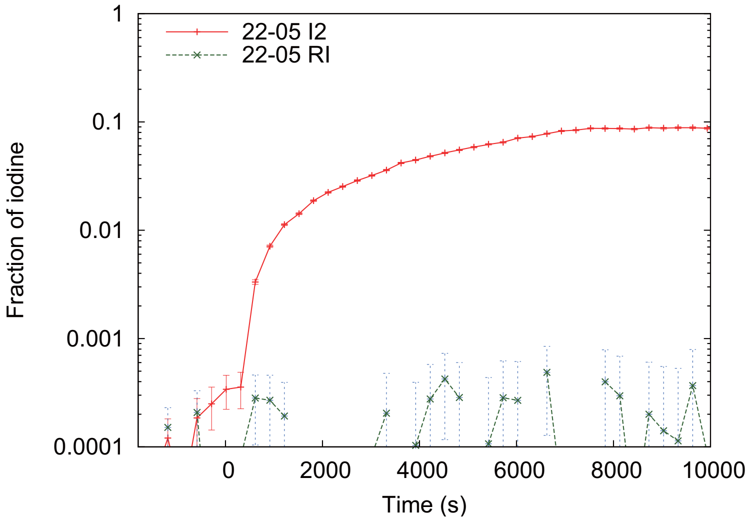




1-5: Ag filters
6-8: AgNO3 filters

Release history





Count efficiency(calibrated)= 0.1970 (Ag sampler)
0.1210 (AgNO3 sampler)

B.2 Cold Experiments

B.2.1 Suppression of Iodine Release by Ammonia Alkaline

Experimental condition

Run No.	**22-C02**
Solution	NH3 0.1M, CsI 1e-4M (ammonia alkaline)
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar
Temperature	24C
Gas	N2 1L/min
Dose rate	0.00 kGy/h
Date	2010/10/18

Irradiation schedule

Stir start	14:45	Ammonia transfer measurement without irradiation.
Sweep start	14:55	
Irrad. start	-	
Irrad. end	-	
Sweep end	16:57	

Solution preparation	Volume(mL)	NH3 conc.(M)	I- conc.(M)
Water	101.000	0.00E+00	0.00E+00
NH3	0.770	1.33E+01	0.00E+00
I-	0.210	0.00E+00	5.00E-02
Vol., conc.	101.980	1.00E-01	1.03E-04

Vessel weight(g)	-		Solution(g)	Loss(g)
with solution	Before(g)	1200.1	-	
	After(g)	1198.0	-	2.100

Chemical measurements

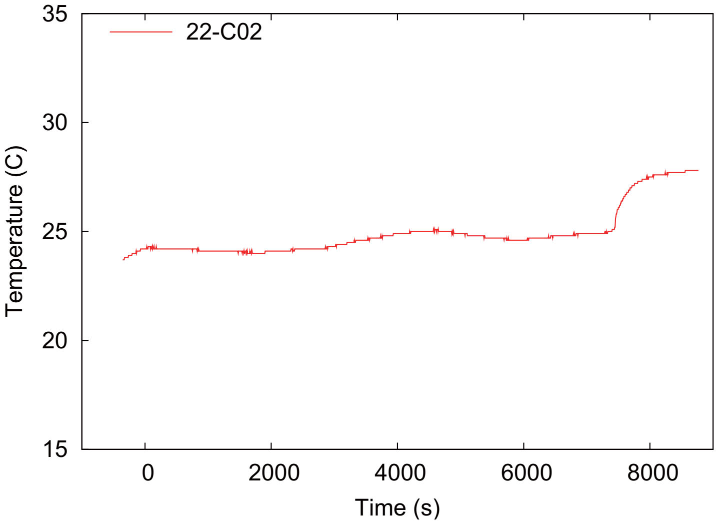
Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
22-C02	11.10	10.81	1.14E-04	1.25E-04	1.07E+00

NH3 conc.		NH3 remained fraction
Before	After	
0.093	0.045	4.75E-01

*Remained fractions: with compensation of liquid decrease due to evaporation.

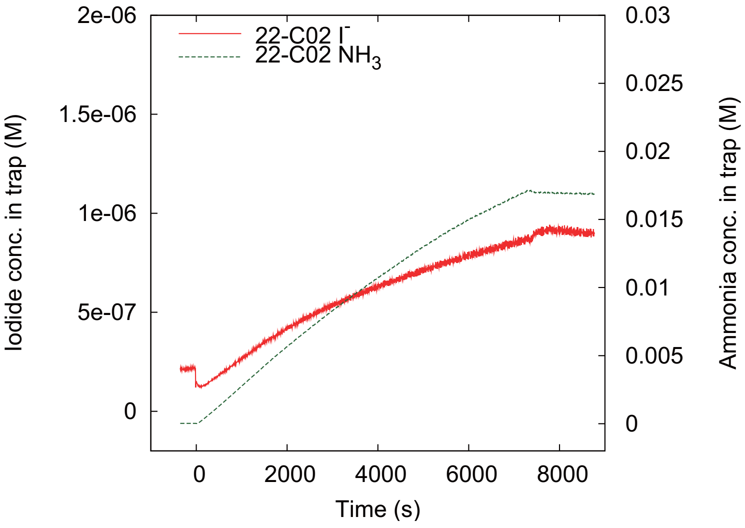
Plots

Temperature	Initial	24 C
	During irrad.	24-25 C



Time 0=start of sweep

Trap I-, NH3 (no correction for NH3 sensitivity of I- probes)



Trap solution 0.1M NaOH
Trap volume 250mL
Trap final conc.(M)

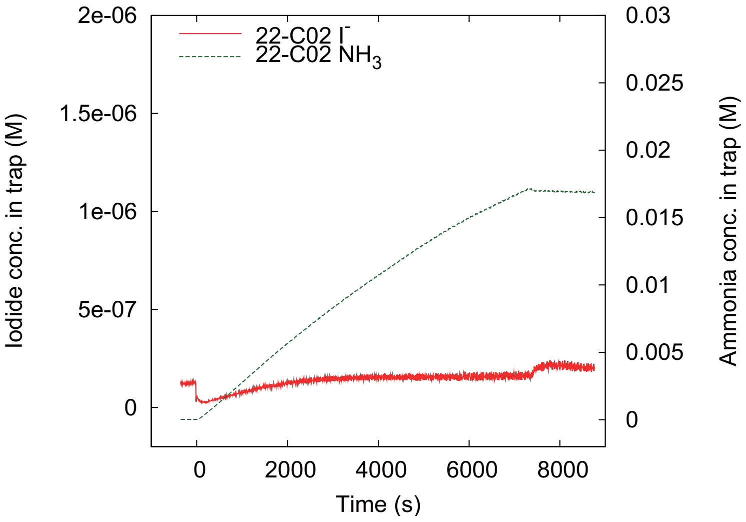
I-	8.68E-07
NH3	1.71E-02

Trapped fractions

I-	1.90E-02
NH3	4.60E-01

*Base=initial measured amount

Trap I-, NH3 (corrected for NH3 sensitivity of I- probes)



Trap solution 0.1M NaOH
Trap volume 250mL
Trap final conc.(M)

I-	1.62E-07
NH3	1.71E-02

Trapped fractions

I-	3.54E-03
NH3	4.60E-01

*Base=initial measured amount

Experimental condition

Run No.	**22-C03**
Solution	NH3 0.1M, CsI 1e-4M (ammonia alkaline)
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar
Temperature	25C
Gas	N2 1L/min
Dose rate	5.08 kGy/h
Date	2010/10/19

Irradiation schedule

Stir start	13:55
Sweep start	13:55
Irrad. start	13:55
Irrad. end	15:55
Sweep end	16:55

Suppression of the iodine release by ammonia alkaline.
Water was swept by N2 for 30min in advance of the exp.
Ammonia and CsI was dissolved after that. (10:44-11:14)
Failed to open the valve at irrad. start. Thus, irrad. was
interrupted and started again (13:57) with the valve open.

Solution preparation	Volume(mL)	NH3 conc.(M)	I- conc.(M)
Water	101.000	0.00E+00	0.00E+00
NH3	0.770	1.33E+01	0.00E+00
I-	0.210	0.00E+00	5.00E-02
Vol., conc.	101.980	1.00E-01	1.03E-04

Vessel weight(g)	1101.0		Solution(g)	Loss(g)
with solution	Before(g)	1200.7	99.7	
	After(g)	1198.6	97.6	2.100

Chemical measurements

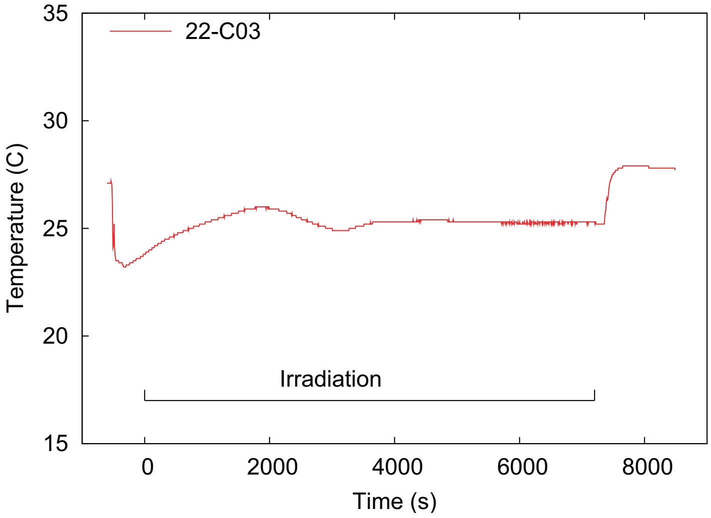
Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
22-C03	11.19	10.89	1.09E-04	1.14E-04	1.02E+00

NH3 conc.		NH3 remained fraction
Before	After	
0.103	0.057	5.44E-01

*Remained fractions: with compensation of liquid decrease due to evaporation.

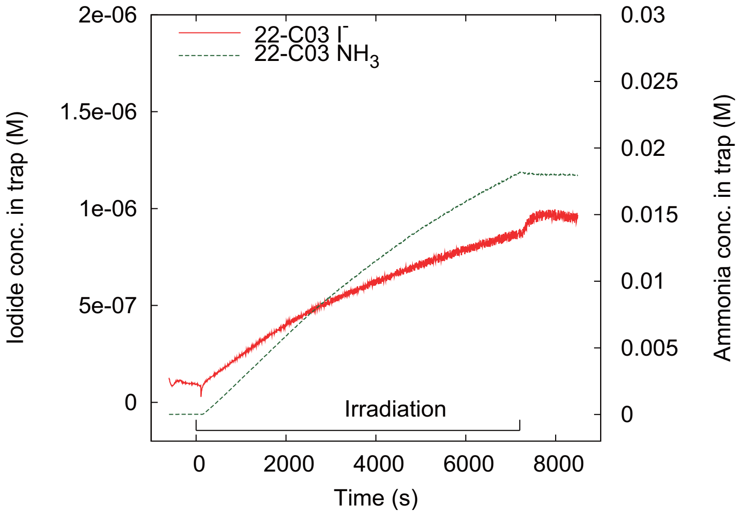
Plots

Temperature	Initial	24 C
	During irrad.	24-26 C



Time 0=start of irrad. and sweep

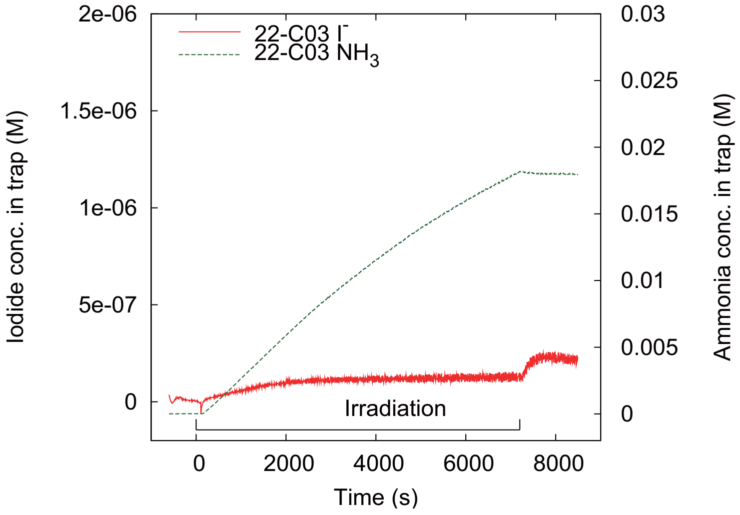
Trap I-, NH3 (no correction for NH3 sensitivity of I- probes)



Trap solution	0.1M NaOH	
Trap volume	250mL	
Trap final conc.(M)		
	I-	8.68E-07
	NH3	1.82E-02
Trapped fractions		
	I-	1.99E-02
	NH3	4.40E-01

*Base=initial measured amount

Trap I-, NH3 (corrected for NH3 sensitivity of I- probes)



Trap solution	0.1M NaOH	
Trap volume	250mL	
Trap final conc.(M)		
	I-	1.24E-07
	NH3	1.82E-02
Trapped fractions		
	I-	2.84E-03
	NH3	4.40E-01

*Base=initial measured amount

Experimental condition

Run No.	**22-C04**
Solution	NH3 0.01M, CsI 1e-4M (ammonia alkaline)
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar
Temperature	25C
Gas	N2 1L/min
Dose rate	5.08 kGy/h
Date	2010/10/20

Irradiation schedule

Stir start	13:09	Suppression of the iodine release by ammonia alkaline.
Sweep start	13:20	NH3 conc. 0.01M
Irrad. start	13:20	Water was swept by N2 for 30min in advance of the exp.
Irrad. end	15:20	Ammonia and CsI was dissolved after that.(11:01-11:31)
Sweep end	15:20	

Solution preparation	Volume(mL)	NH3 conc.(M)	I- conc.(M)
Water	102.000	0.00E+00	0.00E+00
NH3	0.077	1.33E+01	0.00E+00
I-	0.210	0.00E+00	5.00E-02
Vol., conc.	102.287	1.00E-02	1.03E-04

Vessel weight(g)	1100.8		Solution(g)	Loss(g)
with solution	Before(g)	1201.2	100.4	
	After(g)	1198.6	97.8	2.600

Chemical measurements

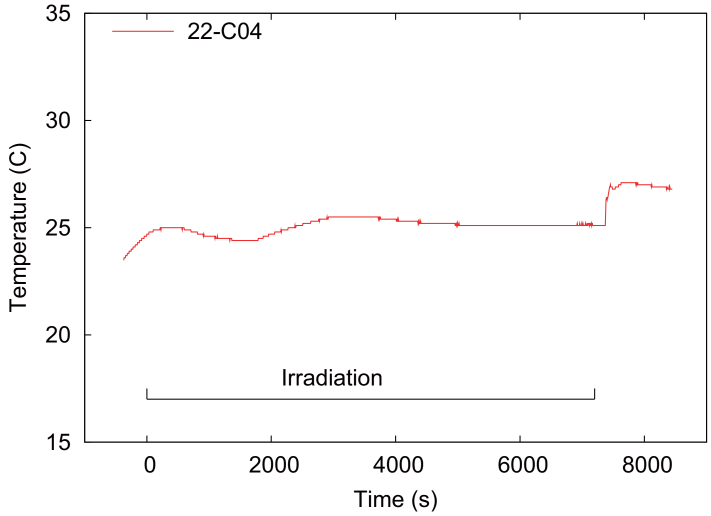
Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
22-C04	10.64	10.28	1.03E-04	1.06E-04	1.00E+00

NH3 conc.		NH3 remained fraction
Before	After	
0.0080	0.0040	4.83E-01

*Remained fractions: with compensation of liquid decrease due to evaporation.

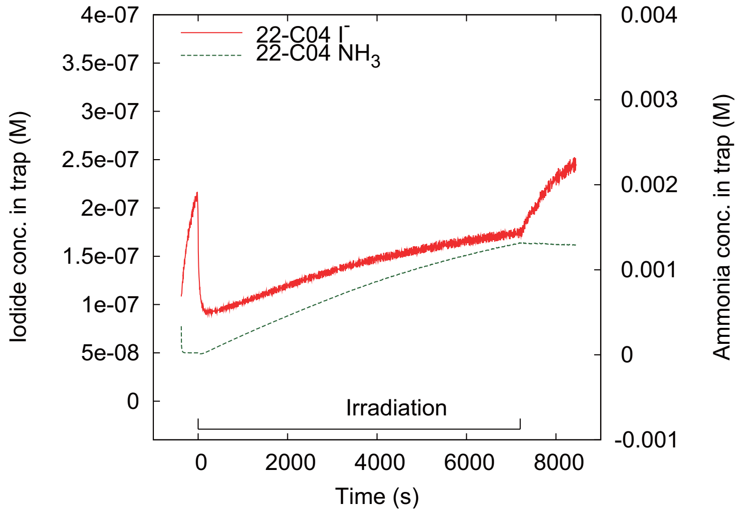
Plots

Temperature	Initial	24 C
	During irrad.	25 C



Time 0=start of irrad. and sweep

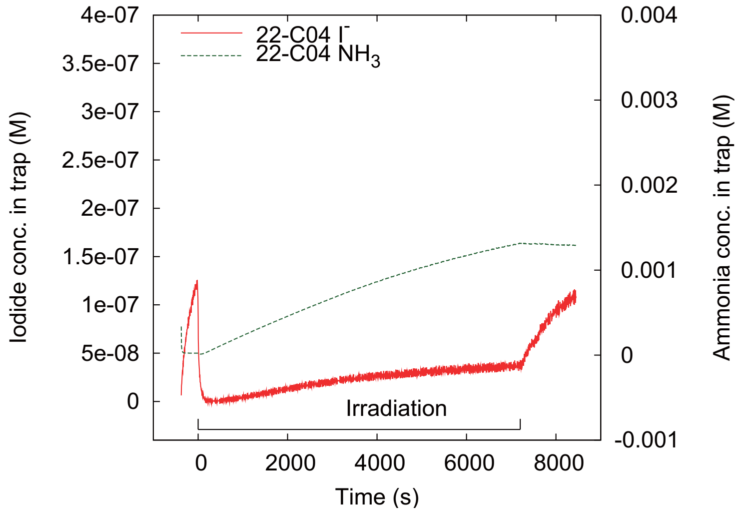
Trap I-, NH3 (no correction for NH3 sensitivity of I- probes)



Trap solution	0.1M NaOH	
Trap volume	250mL	
Trap final conc.(M)		
	I-	1.75E-07
	NH3	1.31E-03
Trapped fractions		
	I-	4.25E-03
	NH3	4.10E-01

*Base=initial measured amount

Trap I-, NH3 (corrected for NH3 sensitivity of I- probes)



Trap solution	0.1M NaOH	
Trap volume	250mL	
Trap final conc.(M)		
	I-	3.67E-08
	NH3	1.31E-03
Trapped fractions		
	I-	8.92E-04
	NH3	4.10E-01

*Base=initial measured amount

Experimental condition

Run No.	**22-C05**	
Solution	NH3 0.1M, CsI 1e-4M (ammonia alkaline)	
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar	
Temperature	25C	
Gas	N2-5%O2 1L/min	
Dose rate	5.07 kGy/h	
Date	2010/10/26	
Irradiation schedule		
	Bypass sweep start	12:26
	Stir start	12:28
	Sweep start	12:30
	Irrad. start	13:00
	Irrad. end	15:00
	Sweep end	15:30

Suppression of the iodine release by ammonia alkaline.
Bypass: gas was let into the trap without sweeping the test vessel.
NH3 conc. 0.1M, 5%O2
No irrad. 30min sweep=>2h irrad.=>No irrad. 30min sweep
The trap I- and NH3 evolution during the irrad and no irrad periods were compaired..

Solution preparation	Volume(mL)	NH3 conc.(M)	I- conc.(M)
Water	101.000	0.00E+00	0.00E+00
NH3	0.770	1.33E+01	0.00E+00
I-	0.210	0.00E+00	5.00E-02
Vol., conc.	101.980	1.00E-01	1.03E-04

Vessel weight(g)	1100.9		Solution(g)	Loss(g)
with solution	Before(g)	1200.2	99.3	
	After(g)	1196.9	96.0	3.300

Chemical measurements

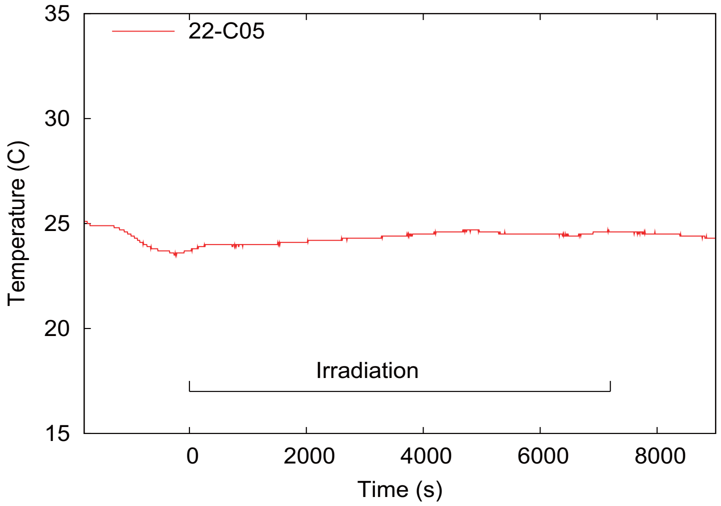
Run No.	pH		I- conc.		I- remained fraction
	Before	After	Before	After	
22-C05	11.24	10.70	1.28E-04	1.24E-04	9.39E-01

NH3 conc.		NH3 remained fraction
Before	After	
0.106	0.040	3.63E-01

*Remained fractions: with compensation of liquid decrease due to evaporation.

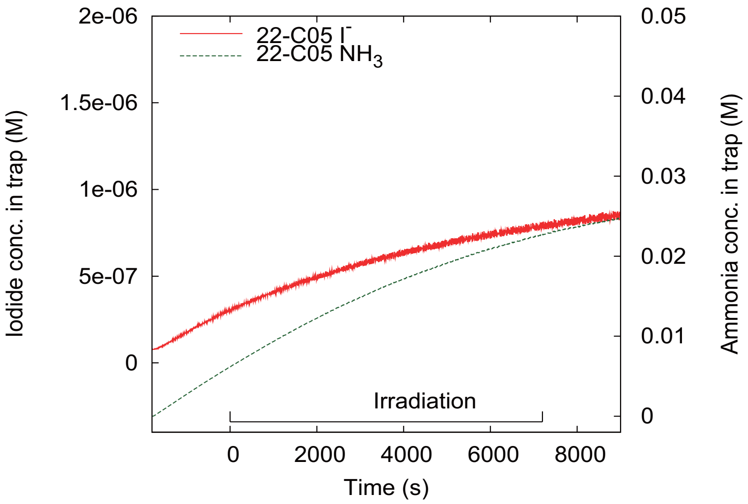
Plots

Temperature	Initial	25 C
	During irrad.	24-25 C



Time 0=start of irrad. and sweep

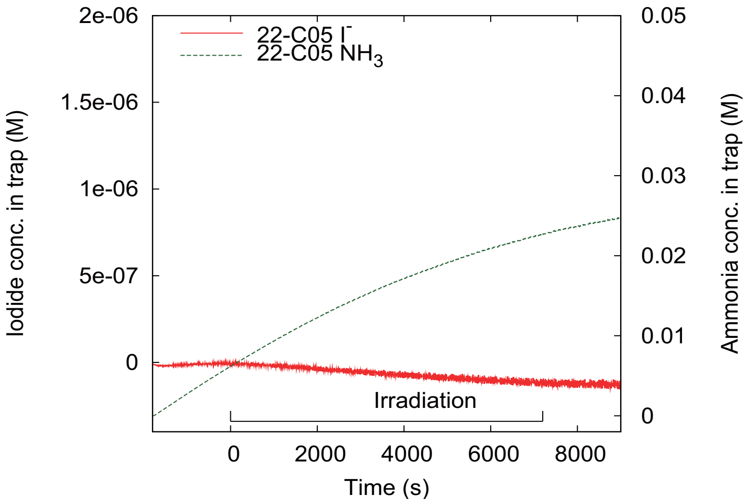
Trap I-, NH3 (no correction for NH3 sensitivity of I- probes)



Trap solution	0.1M NaOH
Trap volume	250mL
Trap final conc.(M)	
I-	8.50E-07
NH3	2.46E-02
Trapped fractions	
I-	1.66E-02
NH3	5.78E-01

*Base=initial measured amount

Trap I-, NH3 (corrected for NH3 sensitivity of I- probes)



Trap solution	0.1M NaOH
Trap volume	250mL
Trap final conc.(M)	
I-	-1.24E-07
NH3	2.46E-02
Trapped fractions	
I-	-2.42E-03
NH3	5.78E-01

*Base=initial measured amount

No significant change was seen between the irrad. and no-irrad. periods.

B.2.2 Comparison of the influence of Organic Compounds: MIBK and Xylene

Experimental condition

Run No.	**22-C10**
Solution	pH=7.0 (Buffer: 0.1M B(OH)3-5.72e-4M NaOH), CsI 1e-4M, o-xylene soaked porous PTFE chip immersec
Volume/Stir	100mL/350rpm-6(dia.)x25 glass stir bar
Temperature	25C
Gas	Air(colosed, gas vol. 286mL)
Dose rate	4.98 kGy/h
Date	2010/12/13

Irradiation schedule

Xylene chip add	13:24
Sample 0	13:24
Sample 1	14:24
Irrad 1 start(0-30min)	14:24
Sample 2	14:54
Irrad 2 start(-60min)	14:55
Sample 3	15:26
Irrad 3 start(-120min)	15:26
Sample 4	16:26
Irrad. end	16:26
30min after	16:56
Sample 5	16:56

Solution preparation	Volume(mL)	I- conc.(M)
Buffer solution	120.000	0.00E+00
I-	0.240	5.00E-02
Vol., conc.	120.240	9.98E-05

Vessel weight(g)	1104.6		Solution(g)	Loss(g)
with solution	Before(g)	1204.3	99.70	
	After(g)	1205.3	99.66	0.039
PTFE chip	Chip (g)	1.0392		
	Chip (dry)(g)	0.9862		
	xylene(g)	0.0530	xylene mol	4.99E-04

Chemical measurements

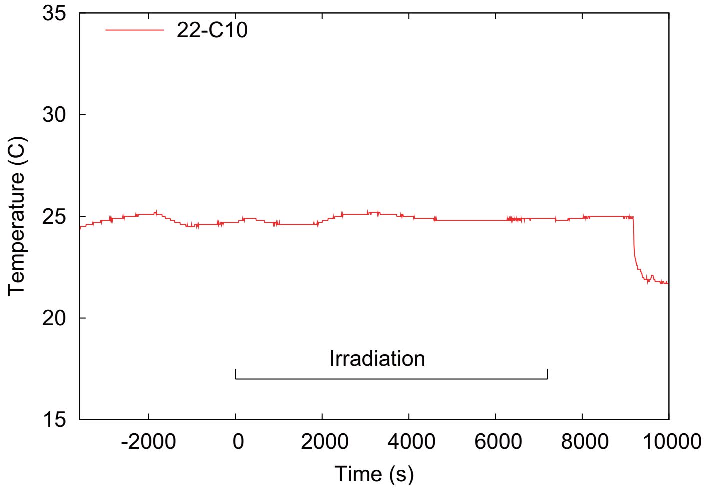
Run No.	pH		I- conc.(M)		I- remained fraction
	Before	After	Before	After	
22-C10	6.91	5.78	1.02E-04	7.14E-05	6.99E-01

GCMS measurements

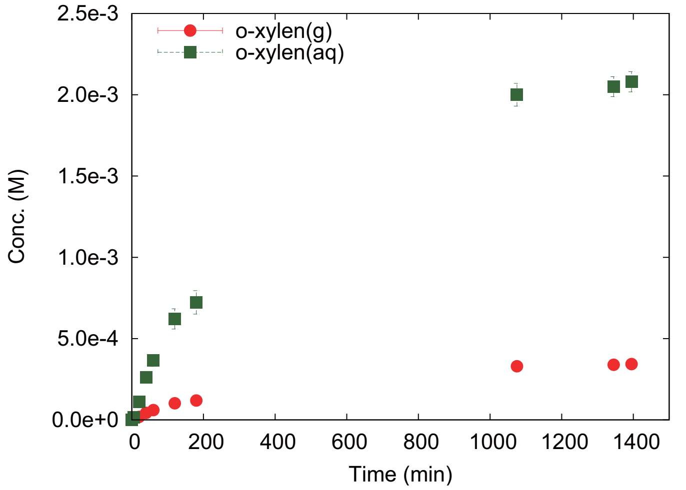
Sample No.	Conc.(M)				Total amount(mol)	
	xylene(gas)	CH3I(gas)	xylene(liq)	CH3I(liq)	xylene	CH3I
0	2.43E-06	0.00E+00	1.47E-05	0.00E+00	2.16E-06	0.00E+00
1	7.66E-05	0.00E+00	4.64E-04	0.00E+00	6.83E-05	0.00E+00
2	6.30E-05	2.71E-08	3.82E-04	1.30E-07	5.62E-05	2.08E-08
3	5.69E-05	4.55E-08	3.45E-04	2.19E-07	5.08E-05	3.49E-08
4	4.95E-05	6.43E-08	3.00E-04	3.10E-07	4.42E-05	4.94E-08
5	7.13E-05	6.27E-08	4.32E-04	3.02E-07	6.36E-05	4.81E-08

Plots

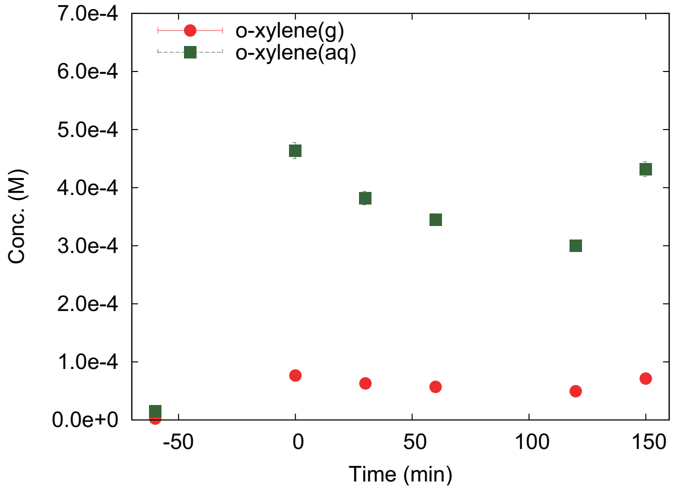
Temperature	Initial	25 C
	During irrad.	25 C



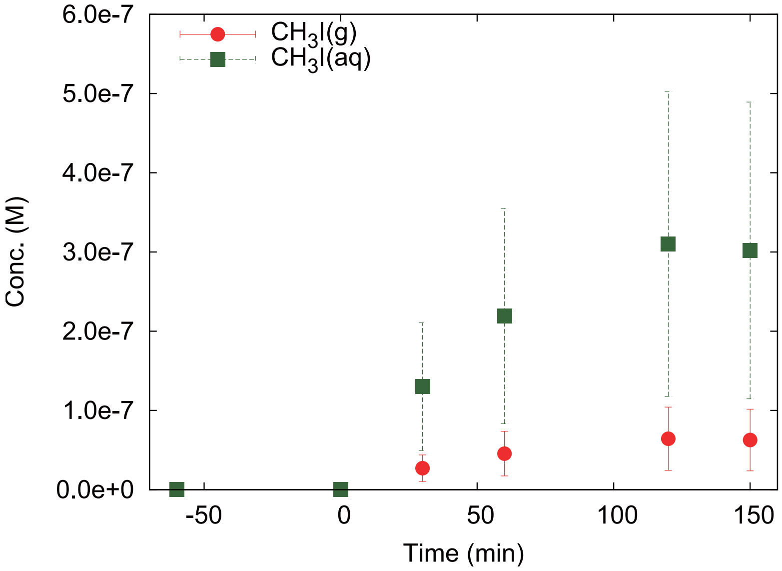
Time 0=start of irrad.



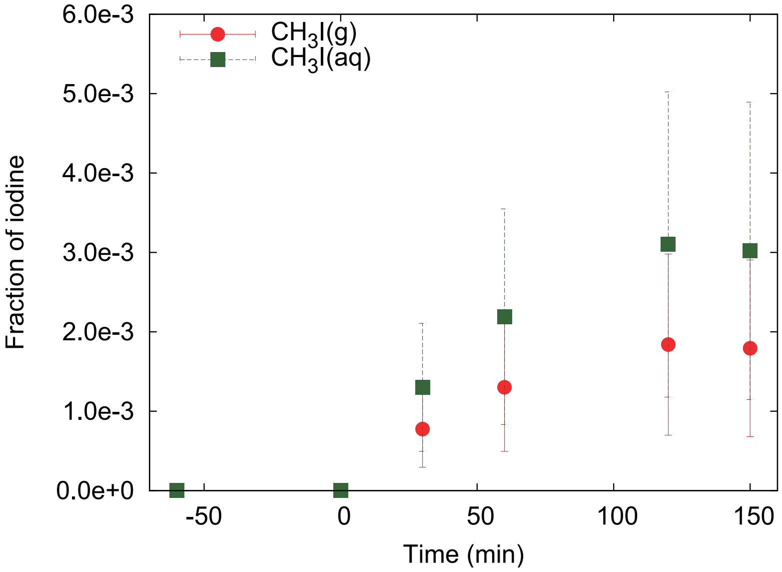
Pre-test
o-xylene soaked porous PTFE chip was immersed in water, and, o-xylene leached was measured. Conc. in liq. was about 4e-4M at 60min.



o-xylene conc. measurement
o-xylene conc. at irrad. start (0s) was comparable to the pre-test conc.

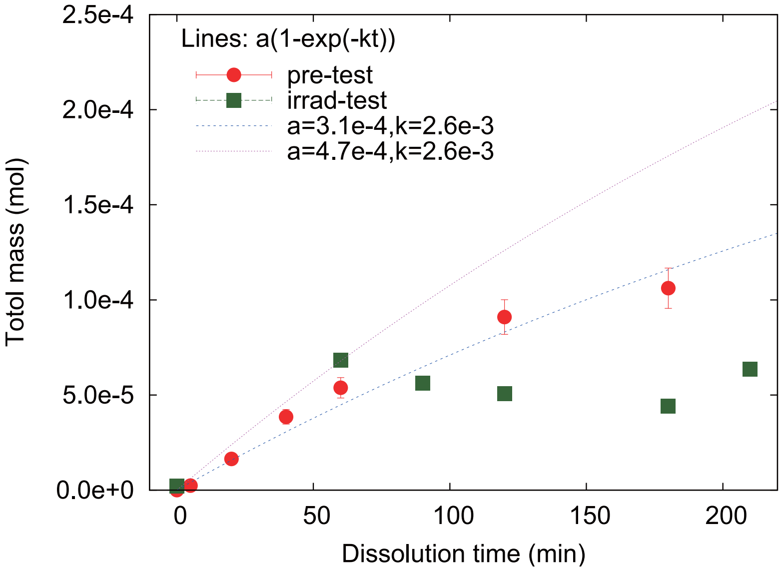


CH3I conc. measurement



CH3I frac. in gas/liq.
(based on initial total I)

o-xylene leaching evaluation (for simulation)
Pre-test leach history was fitted with 1-exp(-kt) function, multiplied by a factor for correction,
so that the o-xylene conc. at the start of irrad. is matched.
M(mol)=4.7e-4 {1-exp(-2.6e-3 t)}, t(min)



Experimental condition

Run No. **22-C11**

Solution pH=7.0 (Buffer: 0.1M B(OH)3-5.72e-4M NaOH), CsI 1e-4M, MIBK 1e-3M added

Volume/Stir 100mL/350rpm-6(dia.)x25 glass stir bar

Temperature 25C

Gas Air(colosed, gas vol. 286mL)

Dose rate 4.98 kGy/h

Date 2010/12/15

Irradiation schedule

MIBK add	13:01
Sample 1	13:02
Irrad 1 start(0-30min)	13:03
Sample 2	13:33
Irrad 2 start(-60min)	13:34
Sample 3	14:04
Irrad 3 start(-120min)	14:05
Sample 4	15:05
Irrad. end	15:05
30min after	15:35
Sample 5	15:35

Solution preparation	Volume(mL)	I- conc.(M)
Buffer solution	120.000	0.00E+00
I-	0.240	5.00E-02
Vol., conc.	120.240	9.98E-05

Vessel weight(g)	926.3		Solution(g)	Loss(g)
with solution	Before(g)	1025.8	99.50	
	After(g)	1026.8	99.50	0
MIBK addition	MIBK sol.(g)	1.0	(1mL)	
	Conc.(M)	0.10	MIBK mol	1.00E-04

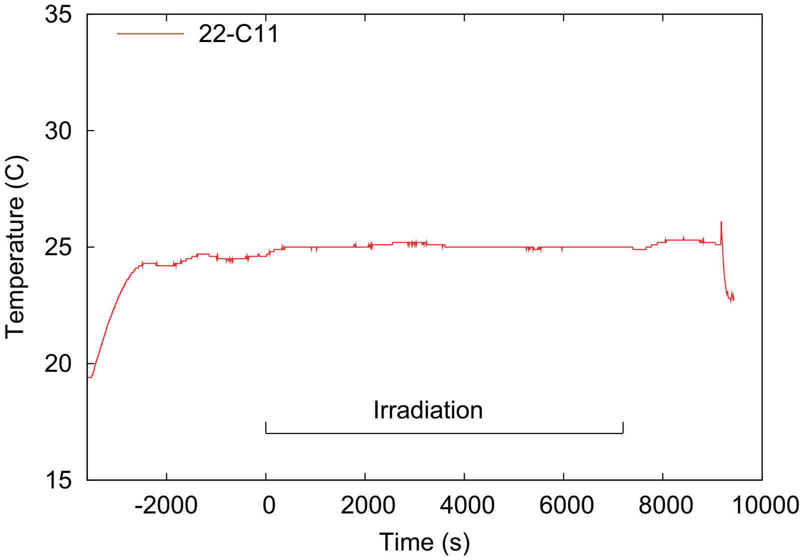
Chemical measurements

Run No.	pH		I- conc.(M)		I- remained fraction
	Before	After	Before	After	
22-C11	6.92	6.20	1.11E-04	9.00E-05	8.10E-01

GCMS measurements

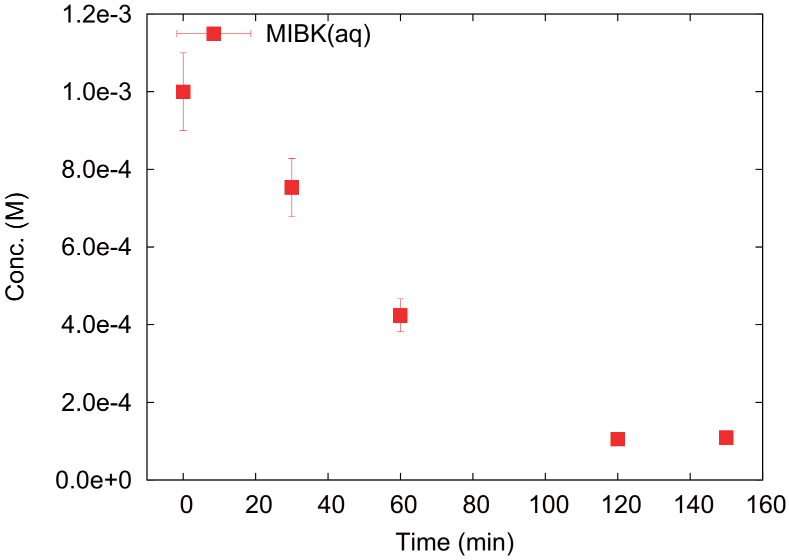
	Conc.(M)				Total amount(mol)	
Sample No.	MIBK(gas)	CH3I(gas)	MIBK(liq)	CH3I(liq)	MIBK	CH3I
1	1.69E-03	0.00E+00	1.00E-03	0.00E+00	5.83E-04	0.00E+00
2	1.27E-03	8.66E-09	7.53E-04	4.17E-08	4.39E-04	6.65E-09
3	7.16E-04	2.95E-08	4.24E-04	1.42E-07	2.47E-04	2.26E-08
4	1.78E-04	7.73E-08	1.05E-04	3.73E-07	6.14E-05	5.94E-08
5	1.84E-04	8.90E-08	1.09E-04	4.29E-07	6.35E-05	6.84E-08

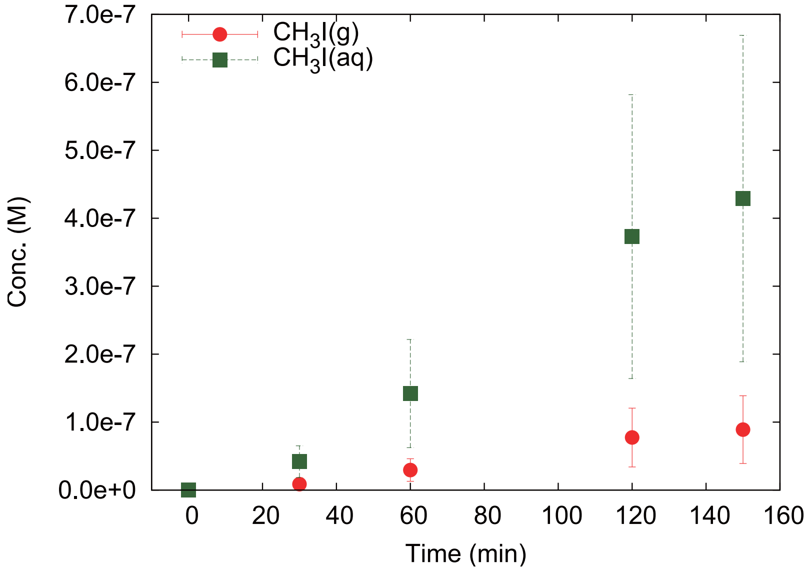
Plots		
Temperature	Initial	25 C
	During irrad.	25 C



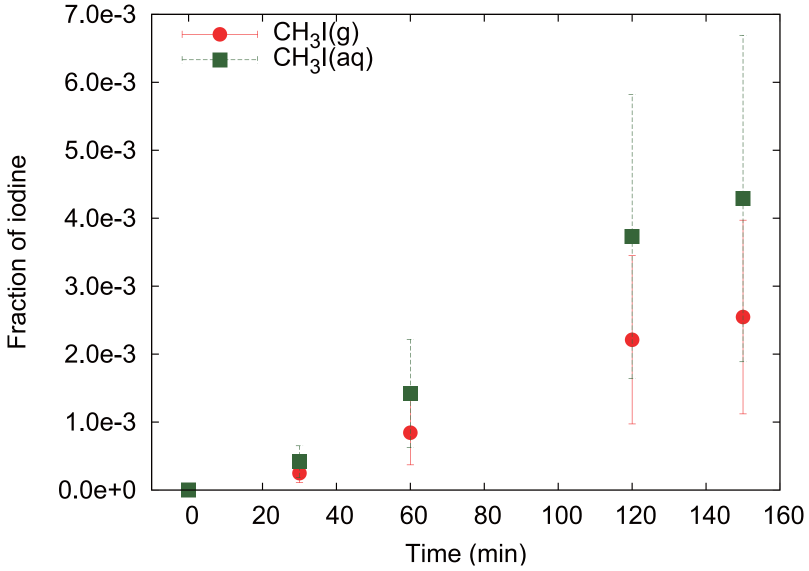
Time 0=start of irrad.

MIBK conc. measurement





CH3I conc. measurement



CH3I frac. in gas/liq.
(based on initial total I)

国際単位系（SI）

表１．SI 基本単位

基本量	SI 基本単位	
	名称	記号
長さ	メートル	m
質量	キログラム	kg
時間	秒	s
電流	アンペア	A
熱力学温度	ケルビン	K
物質量	モル	mol
光度	カンデラ	cd

表２．基本単位を用いて表されるSI組立単位の例

組立量	SI 基本単位	
	名称	記号
面積	平方メートル	m^{2}
体積	立法メートル	m^{3}
速さ，速度	メートル毎秒	m/s
加速度	メートル毎秒毎秒	m/s^{2}
波数	毎メートル	m^{-1}
密度，質量密度	キログラム毎立方メートル	kg/m^{3}
面積密度	キログラム毎平方メートル	kg/m^{2}
比体積	立方メートル毎キログラム	m^{3}/kg
電流密度	アンペア毎平方メートル	A/m^{2}
磁界の強さ	アンペア毎メートル	A/m
量濃度(a)，濃度	モル毎立方メートル	mol/m^{3}
質量濃度	キログラム毎立法メートル	kg/m^{3}
輝度	カンデラ毎平方メートル	cd/m^{2}
屈折率(b)	（数字の）　1	1
比透磁率(b)	（数字の）　1	1

(a)　量濃度（amount concentration）は臨床化学の分野では物質濃度（substance concentration）ともよばれる。

(b)　これらは無次元量あるいは次元１をもつ量であるが、そのことを表す単位記号である数字の１は通常は表記しない。

表３．固有の名称と記号で表されるSI組立単位

組立量	SI 組立単位			
	名称	記号	他のSI単位による表し方	SI基本単位による表し方
平面角	ラジアン(b)	rad	1 (b)	m/m
立体角	ステラジアン(b)	sr(c)	1 (b)	m$^{2/}$m^{2}
周波数	ヘルツ(d)	Hz		s^{-1}
力	ニュートン	N		m kg s^{-2}
圧力，応力	パスカル	Pa	N/m^{2}	m^{-1} kg s^{-2}
エネルギー，仕事，熱量	ジュール	J	N m	m^{2} kg s^{-2}
仕事率，工率，放射束	ワット	W	J/s	m^{2} kg s^{-3}
電荷，電気量	クーロン	C		s A
電位差（電圧），起電力	ボルト	V	W/A	m^{2} kg s^{-3} A^{-1}
静電容量	ファラド	F	C/V	m^{-2} kg^{-1} s^{4} A^{2}
電気抵抗	オーム	Ω	V/A	m^{2} kg s^{-3} A^{-2}
コンダクタンス	ジーメンス	S	A/V	m^{-2} kg^{-1} s^{3} A^{2}
磁束	ウエーバ	Wb	Vs	m^{2} kg s^{-2} A^{-1}
磁束密度	テスラ	T	Wb/m^{2}	kg s^{-2} A^{-1}
インダクタンス	ヘンリー	H	Wb/A	m^{2} kg s^{-2} A^{-2}
セルシウス温度	セルシウス度(e)	℃		K
光束	ルーメン	lm	cd sr(c)	cd
照度	ルクス	lx	lm/m^{2}	m^{-2} cd
放射性核種の放射能(f)	ベクレル(d)	Bq		s^{-1}
吸収線量, 比エネルギー分与, カーマ	グレイ	Gy	J/kg	m^{2} s^{-2}
線量当量, 周辺線量当量, 方向性線量当量, 個人線量当量	シーベルト(g)	Sv	J/kg	m^{2} s^{-2}
酸素活性	カタール	kat		s^{-1} mol

(a)SI接頭語は固有の名称と記号を持つ組立単位と組み合わせても使用できる。しかし接頭語を付した単位はもはやコヒーレントではない。

(b)ラジアンとステラジアンは数字の１に対する単位の特別な名称で、量についての情報をつたえるために使われる。実際には、使用する時には記号rad及びsrが用いられるが、習慣として組立単位としての記号である数字の１は明示されない。

(c)測光学ではステラジアンという名称と記号srを単位の表し方の中に、そのまま維持している。

(d)ヘルツは周期現象についてのみ、ベクレルは放射性核種の統計的過程についてのみ使用される。

(e)セルシウス度はケルビンの特別な名称で、セルシウス温度を表すために使用される。セルシウス度とケルビンの単位の大きさは同一である。したがって、温度差や温度間隔を表す数値はどちらの単位で表しても同じである。

(f)放射性核種の放射能（activity referred to a radionuclide）は、しばしば誤った用語で”radioactivity”と記される。

(g)単位シーベルト（PV,2002,70,205）についてはCIPM勧告2（CI-2002）を参照。

表４．単位の中に固有の名称と記号を含むSI組立単位の例

組立量	SI 組立単位		
	名称	記号	SI 基本単位による表し方
粘度	パスカル秒	Pa s	m^{-1} kg s^{-1}
力のモーメント	ニュートンメートル	N m	m^{2} kg s^{-2}
表面張力	ニュートン毎メートル	N/m	kg s^{-2}
角速度	ラジアン毎秒	rad/s	m m^{-1} s^{-1}=s^{-1}
角加速度	ラジアン毎秒毎秒	rad/s^{2}	m m^{-1} s^{-2}=s^{-2}
熱流密度，放射照度	ワット毎平方メートル	W/m^{2}	kg s^{-3}
熱容量，エントロピー	ジュール毎ケルビン	J/K	m^{2} kg s^{-2} K^{-1}
比熱容量，比エントロピー	ジュール毎キログラム毎ケルビン	J/(kg K)	m^{2} s^{-2} K^{-1}
比エネルギー	ジュール毎キログラム	J/kg	m^{2} s^{-2}
熱伝導率	ワット毎メートル毎ケルビン	W/(m K)	m kg s^{-3} K^{-1}
体積エネルギー	ジュール毎立方メートル	J/m^{3}	m^{-1} kg s^{-2}
電界の強さ	ボルト毎メートル	V/m	m kg s^{-3} A^{-1}
電荷密度	クーロン毎立方メートル	C/m^{3}	m^{-3} sA
表面電荷	クーロン毎平方メートル	C/m^{2}	m^{-2} sA
電束密度，電気変位	クーロン毎平方メートル	C/m^{2}	m^{-2} sA
誘電率	ファラド毎メートル	F/m	m^{-3} kg^{-1} s^{4} A^{2}
透磁率	ヘンリー毎メートル	H/m	m kg s^{-2} A^{-2}
モルエネルギー	ジュール毎モル	J/mol	m^{2} kg s^{-2} mol^{-1}
モルエントロピー, モル熱容量	ジュール毎モル毎ケルビン	J/(mol K)	m^{2} kg s^{-2} K^{-1} mol^{-1}
照射線量（Ｘ線及びγ線）	クーロン毎キログラム	C/kg	kg^{-1} sA
吸収線量率	グレイ毎秒	Gy/s	m^{2} s^{-3}
放射強度	ワット毎ステラジアン	W/sr	m^{4} m^{-2} kg s^{-3}=m^{2} kg s^{-3}
放射輝度	ワット毎平方メートル毎ステラジアン	W/(m^{2} sr)	m^{2} m^{-2} kg s^{-3}=kg s^{-3}
酵素活性濃度	カタール毎立方メートル	kat/m^{3}	m^{-3} s^{-1} mol

表５．SI 接頭語

乗数	接頭語	記号	乗数	接頭語	記号
10^{24}	ヨタ	Y	10^{-1}	デシ	d
10^{21}	ゼタ	Z	10^{-2}	センチ	c
10^{18}	エクサ	E	10^{-3}	ミリ	m
10^{15}	ペタ	P	10^{-6}	マイクロ	µ
10^{12}	テラ	T	10^{-9}	ナノ	n
10^{9}	ギガ	G	10^{-12}	ピコ	p
10^{6}	メガ	M	10^{-15}	フェムト	f
10^{3}	キロ	k	10^{-18}	アト	a
10^{2}	ヘクト	h	10^{-21}	ゼプト	z
10^{1}	デカ	da	10^{-24}	ヨクト	y

表６．SIに属さないが、SIと併用される単位

名称	記号	SI 単位による値
分	min	1 min=60s
時	h	1h =60 min=3600 s
日	d	1 d=24 h=86 400 s
度	°	1°=(π/180) rad
分	’	1’=(1/60)°=(π/10800) rad
秒	”	1”=(1/60)’=(π/648000) rad
ヘクタール	ha	1ha=1hm^{2}=10^{4}m^{2}
リットル	L，l	1L=1l=1dm^{3}=10^{3}cm^{3}=10^{-3}m^{3}
トン	t	1t=10^{3} kg

表７．SIに属さないが、SIと併用される単位で、SI単位で表される数値が実験的に得られるもの

名称	記号	SI 単位で表される数値
電子ボルト	eV	1eV=1.602 176 53(14)×10^{-19}J
ダルトン	Da	1Da=1.660 538 86(28)×10^{-27}kg
統一原子質量単位	u	1u=1 Da
天文単位	ua	1ua=1.495 978 706 91(6)×10^{11}m

表８．SIに属さないが、SIと併用されるその他の単位

名称	記号	SI 単位で表される数値
バール	bar	1 bar=0.1MPa=100kPa=10^{5}Pa
水銀柱ミリメートル	mmHg	1mmHg=133.322Pa
オングストローム	Å	1 Å=0.1nm=100pm=10^{-10}m
海里	M	1 M=1852m
バーン	b	1 b=100fm^{2}=(10^{-12}cm)2=10^{-28}m^{2}
ノット	kn	1 kn=(1852/3600)m/s
ネーパ	Np	SI単位との数値的な関係は、対数量の定義に依存。
ベル	B	
デジベル	dB	

表９．固有の名称をもつCGS組立単位

名称	記号	SI 単位で表される数値
エルグ	erg	1 erg=10^{-7} J
ダイン	dyn	1 dyn=10^{-5}N
ポアズ	P	1 P=1 dyn s cm^{-2}=0.1Pa s
ストークス	St	1 St =1cm^{2} s^{-1}=10^{-4}m^{2} s^{-1}
スチルブ	sb	1 sb =1cd cm^{-2}=10^{4}cd m^{-2}
フォト	ph	1 ph=1cd sr cm^{-2} 10^{4}lx
ガル	Gal	1 Gal =1cm s^{-2}=10^{-2}ms^{-2}
マクスウェル	Mx	1 Mx = 1G cm^{2}=10^{-8}Wb
ガウス	G	1 G =1Mx cm^{-2} =10^{-4}T
エルステッド(c)	Oe	1 Oe ≙ (10^{3}/4π)A m^{-1}

(c)　３元系のCGS単位系とSIでは直接比較できないため、等号「 ≙ 」は対応関係を示すものである。

表10．SIに属さないその他の単位の例

名称	記号	SI 単位で表される数値
キュリー	Ci	1 Ci=3.7×10^{10}Bq
レントゲン	R	1 R = 2.58×10^{-4}C/kg
ラド	rad	1 rad=1cGy=10^{-2}Gy
レム	rem	1 rem=1 cSv=10^{-2}Sv
ガンマ	γ	1 γ =1 nT=10-9T
フェルミ		1フェルミ=1 fm=10-15m
メートル系カラット		1メートル系カラット = 200 mg = 2×10-4kg
トル	Torr	1 Torr = (101 325/760) Pa
標準大気圧	atm	1 atm = 101 325 Pa
カロリー	cal	1cal=4.1858J（「15℃」カロリー），4.1868J（「IT」カロリー）4.184J（「熱化学」カロリー）
ミクロン	µ	1 µ =1µm=10^{-6}m

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