

Radioiodine Chemistry: The Unfinished Story

J.C. Wren

Department of Chemistry, University of Western Ontario

London, Ontario, CANADA

jcwren@uwo.ca

ABSTRACT

Because of its importance in the source term for postulated nuclear accidents, considerable effort has been expended worldwide to understand the behaviour of radioiodine under accident conditions. This has led to significant progress in our understanding of complex and sophisticated radiolysis-driven iodine chemistry and transport behaviour under potential reactor accident conditions. Models that describe, with some confidence, the chemistry of radioiodines and, in particular, the chemistry of those radioiodine species having a propensity to become airborne under appropriate conditions, have been developed and used to establish and prioritize key processes and phenomena under conditions of interest.

The focus of research can now shift to address the more challenging roles that other chemical species and chemically reactive surfaces can play in influencing radioiodine chemistry. In the real world, we face situations in which many chemicals may be present (from paints, solvents, etc) as 'impurities' in the environment, and these additional chemicals have the potential to strongly influence radioiodine chemistry. This is the most evident through the reactions of organic compounds with water radiolysis products and consequential impact of these reactions on radioiodine species in a solution. Heterogeneous reactions of radioiodine with metallic surfaces raise the level of complexity further: metallic surfaces may release metal ions that could react catalytically with radiolysis products, and, more importantly, such surfaces could act as temporary or permanent radioiodine sinks. This becomes a dominant concern when trying to control, monitor, or prevent airborne radioiodine movement.

This paper will briefly summarize the successes that have been achieved by the international nuclear radioiodine chemistry community and then explore the challenges and opportunities that are now being explored.

1. INTRODUCTION

Iodine has been a long-standing concern in safety analysis because of its radiological significance, combined with its chemical and physical properties (particularly its volatility at low temperatures), its relative abundance in irradiated fuel, and the large fractional releases from fuel predicted for it during accidents. The kinetics of key chemical and transport processes influencing iodine behaviour under accident conditions are rather complex, and have been the subject of extensive studies by the worldwide nuclear community for more than three decades. In that time, and particularly over the past 10 years, these efforts have generated a large body of knowledge in diverse areas and have led to significant progress in our understanding of complex and sophisticated radiolysis-driven iodine chemistry and transport behaviour under potential reactor accident conditions.

Much of the knowledge has been summarized in the status report on iodine chemistry prepared by a group of expert for the OECD [1], and previously by Klausmann [2] and by Wren et al. [3]. These reports provide an overview of the important facets of iodine

chemistry¹. The topic of this paper is limited to a discussion of iodine behaviour in the containment building during the 'chemistry' phase of an accident scenario. This is the period during which the chemical state of fission product radioiodine and the distribution of radioiodine species between the different phases are evolving rapidly towards a steady state under the influence of radiation.

Models that describe, with some confidence, the chemistry of radioiodines and, in particular, the chemistry of those radioiodine species having a propensity to become airborne under appropriate conditions, have been developed and used to establish and prioritize key processes and phenomena under conditions of interest. However, the international community has not yet agreed on a common approach to modeling the chemistry, and the development of an associated safety analysis tool with the capability of predicting iodine behaviour, with confidence, in a 'real' system. This is evidenced by the spread in the results of exercises on iodine modeling sponsored by CSNI (International Standard Problems (ISP) 41 and 46).

The ISP exercises do not reflect uncertainty in the accuracy or adequacy of the fundamental reaction chemistry data for iodine species, particularly in the aqueous phase. Rather the different results stem from the more difficult challenge of how to model the influence of the 'real' environment on the chemical evolution of the iodine. Real here refers to non-ideal aqueous or gaseous chemistry in an actual containment building with minor chemical constituents (impurities) and surfaces (either fixed or particulate) that can play a role in the iodine chemistry. Defining the real environment appropriately, and necessarily through some level of approximation, is not an easy task.

As a result of our progress on the basic iodine chemistry, the focus of research can now shift to address the more challenging problem of predicting the influence of the minor chemical species and chemically reactive surfaces on radioiodine chemistry. The importance of additional chemicals in water is the most evident through the reactions of organic compounds (coming from paints, solvents, etc.) with water radiolysis products and consequential impact of these reactions on radioiodine species in a solution. Heterogeneous reactions of radioiodine with metallic surfaces raise the level of complexity further: metallic surfaces may release metal ions that could react catalytically with radiolysis products, and, more importantly, such surfaces could act as temporary or permanent radioiodine sinks. This becomes a dominant concern when trying to control, monitor, or prevent airborne radioiodine movement.

The challenge of developing our predictive chemistry tools to deal with more appropriate representations of the potential accident environment provides us with a mandate for focussing the next stage of international work. A common focus is required because the wide range of potential reaction partners influencing iodine chemistry could be overwhelming and multiple, parallel attempts to develop simplifying approximations could lead a situation where disagreement between predictive tools becomes confused and difficult to disentangle. A common agreement on the best route forward, the most important secondary chemical processes and the priority increments to our predictive tools offers the best opportunity to develop consistent and convergent predictive capabilities. However, even within a common approach there should still be room for individual exploration of separate effects so that we, as a community, do not lose credibility by sustaining a critical approach to safety analysis tool development.

¹ These reports provide extensive references, which will not be repeated in this paper.

Before addressing the particular initiatives that are being carried out to address this challenging situation, it is helpful to briefly review the status of the basic iodine chemistry knowledge.

2. THE CURRENT STATE OF IODINE CHEMISTRY

2.1 Background

The impetus for developing predictive tools for modelling of radioiodine behaviour arose from the events that occurred during the accident at Three Mile Island Unit 2 (TMI-2). During that accident, that resulted in severe damage to the fuel in the core, an extremely small fraction of the core radioiodine was found airborne (and less released). This was in sharp contrast to the licensing assumptions regarding the behaviour of radioiodine. Based on limited detailed knowledge of accident progressions and limited qualified safety analysis tools, 'conservative' assumptions were made regarding radioiodine behaviour. The US NRC TID 18444 established a set of licensing basis assumptions for an accident like that which occurred at TMI-2 [4]. These included:

- 50% of the core inventory of iodine is instantaneously released to containment,
- 50% of the iodine in containment is deposited onto surfaces rapidly, and
- The distribution of iodine entering containment was considered to be: 91% I₂, 5% aerosol particulates, and 4% volatile organic iodides.

The actual situation at TMI-2 showed that these early assumptions were incorrect and that reliance on them could lead to inappropriate safety design decisions and inappropriate emergency management plans and provisions. The TMI-2 event triggered a large effort to understand the progression of beyond design basis accidents, a significant component of which was an effort to improve the understanding of iodine behaviour. One result of that effort was the establishment in the United States of a new methodology for predicting source terms. The new treatment was documented in US NUREG-1465 which updated the assumption for iodine speciation in containment as: 95% CsI, 5% I₂, and 0.15% volatile organic iodides [5].

The recommendations of NUREG-1465 were based largely on research data available in 1990. This treatment still contains substantial conservatism (e.g., assumption that 5% of the iodine is present as I₂) and there is considerable opportunity to further improve source term predictions with a technically sound basis for treating the behaviour of iodine in an accident.

2.2 Containment Iodine Chemistry and Transport

It is generally accepted that, under most accident conditions, iodine would be released from fuel and reactor core into the containment atmosphere primarily as water-soluble iodide salts such as CsI. Thus, once released into containment, radioiodine would quickly dissolve as non-volatile I⁻ in water originating from the discharged coolant and the operation of any spray or dousing systems. However, in the presence of high radiation fields in a post-accident containment, I⁻ could be oxidized through radiolytic driven processes to form volatile molecular iodine (I₂). Molecular iodine in the aqueous phase would partition into the gas phase, be further oxidized to other non-volatile iodine oxides (such as IO₃⁻), or be reduced back to its original non-volatile I⁻ state by thermal or radiolytic processes. Iodine species in solution could also react with organic compounds present to form organic iodides, that could subsequently decompose to I⁻ or, like I₂, partition into the gas phase. Finally, the deposition of both gaseous and aqueous iodine species on surfaces could alter the concentrations of iodine species or induce

surface-catalyzed transformations. These processes are schematically shown in Figure 1. The relative rates of these processes determine the gaseous iodine concentration, at any particular time, in containment following an accident.

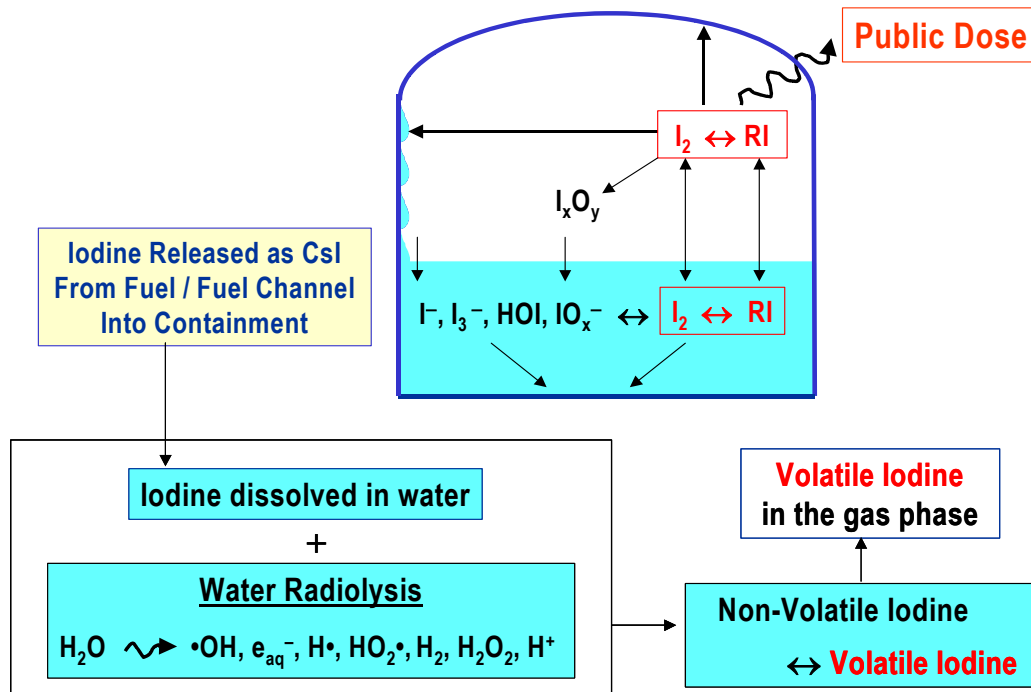


Figure 1: Schematic of Iodine Chemistry and Transport in Containment

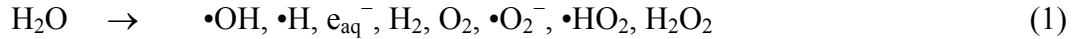
The iodine system in the post-accident containment does not reach thermal equilibrium because reactive chemicals are continuously produced by radiolysis and the temperature in containment would be relatively low ($<150^\circ C$). Hence, chemical thermodynamic calculations are not sufficient to determine iodine speciation and the kinetics of individual reaction paths, not just the final stable chemical state, must be considered in predicting the volatile iodine fraction. However, the dominant reactions may occur fast enough so that the system may reach steady state, which can simplify the modelling of iodine behaviour.

Determination of the rates of all the chemical processes under the post-accident containment conditions is rather complicated. The primary factors contributing to this complication are (a) the strong coupling between the chemistry in the aqueous and gas phases (homogeneous reactions), surface interactions (heterogeneous reactions) and mass transport, and (b) the complex chemistry, both in the homogenous phases and on surfaces, in the presence of highly ionizing radiation.

The complex chemistry arises because under the post-accident conditions, iodine could exist in various oxidation states, ranging from -1 to $+7$ (such as I^- , RI, I_2 , HOI, IO_2^- , IO_3^- and IO_4^- , where RI represents organic iodine). The chemical reactivities and transport properties of different species vary considerably (several orders of magnitude in rate constants), and only some of these iodine species (RI and I_2) are volatile. To determine

iodine volatility in containment, iodine speciation and transport as a function of time must be considered.

An ionizing radiation environment in the post-accident containment will cause the decomposition of water to produce chemically reactive species:



Because iodine would be present in containment water at a trace level, the conversion between non-volatile to volatile iodine species would occur mainly via reactions with water radiolysis products. This complicates the iodine reaction kinetics, because the individual reactions of iodine species and radiolysis products, and the concentrations of all the radiolysis products as well as iodine species, need to be followed. Thus, for example, for iodine species, $(\text{IOD})_n$:

$$\frac{d[(\text{IOD})_n]}{dt} = \sum_{\ell \neq n} \sum_m k_{\text{PROD}-n}^{\ell m} \cdot [(\text{IOD})_\ell] \cdot [(\text{RADP})_m] - \sum_m k_{\text{DEC}-n}^m \cdot [(\text{IOD})_n] \cdot [(\text{RADP})_m] \quad (2)$$

where $k_{\text{PROD}-n}^{\ell m}$ is the rate constant for the reaction of iodine species ℓ , $(\text{IOD})_\ell$ and water radiolysis product m , $(\text{RADP})_m$ to form $(\text{IOD})_n$; $k_{\text{DEC}-n}^m$ is the decomposition rate constant of $(\text{IOD})_n$ by $(\text{RADP})_m$. We must solve the coupled rate equations for all iodine species, and the more iodine oxidation states, the more coupled differential equations and, of course, this includes the thermal iodine reactions that are fast (e.g., iodine hydrolysis which converts I_2 to non-volatile iodide and iodate).

Water radiolysis products not only react with each other, but with other chemicals that may be present, in addition to iodine species. Even in the absence of any impurities, the reaction kinetics of water radiolysis products is a complex problem, due to the large number of the relevant reactions:

$$\frac{d[(\text{RADP})_n]}{dt} = k_{\text{RAD}-\text{PROD}}^n \cdot D_R - \sum_{m \neq n} k_{\text{RAD}-\text{DEC}-n}^m \cdot [(\text{RADP})_n] \cdot [(\text{RADP})_m] \quad (3)$$

where $k_{\text{RAD}-\text{PROD}}^n$ is the rate constant for the production of water radiolysis product n , $(\text{RADP})_n$, by absorption of radiation, D_R is the radiation absorption dose rate by water, and $k_{\text{RAD}-\text{DEC}-n}^m$ is the decomposition rate constant of $(\text{RADP})_n$ by other radiolysis product $(\text{RADP})_m$.

Some dissolved impurities, such as organic compounds and trace metal ions will react rapidly with the radiolysis products. This further increases the number of reactions to be included in the rate equations in order to keep track of the concentrations of water radiolysis products. As well these radiolytic reactions of impurities can also affect the chemical environment (e.g., through changing the water pH). For example, organic impurities react to form organic acids and finally to CO_2 . This change in environment could alter the rate constant, k of other individual reactions that depend on the pH. During the radiolytic decomposition, organic reaction intermediates may be particularly reactive with iodine species (mainly I_2) leading to the formation of a range of organic iodides. Because of the importance of the levels of impurities in rate determination, the sources of these impurities and their dissolution rates into the bulk water phase are additional factors to consider in evaluating iodine volatility under accident conditions. The key aqueous chemical processes are summarized schematically in Figure 2.

Steam or water vapour in the gas phase would also absorb radiation and decompose, creating radiolysis products that can undergo similar reactions, although at much different rates, with each other, gaseous iodine species (I_2 and organic iodides) and airborne contaminants (e.g., gaseous organic compounds) (Figure 3). The radiolysis of humid air also generates ozone (O_3) and NO_x . The formation of O_3 could increase the conversion of I_2 to non-volatile I_xO_y , whereas the formation of NO_x could continuously introduce nitrate and nitrite ions into water elsewhere in containment. The presence of nitrite and nitrate (i.e., as nitric acid) would not only lower the pH of this water but also would react with water radiolysis products semi-catalytically.

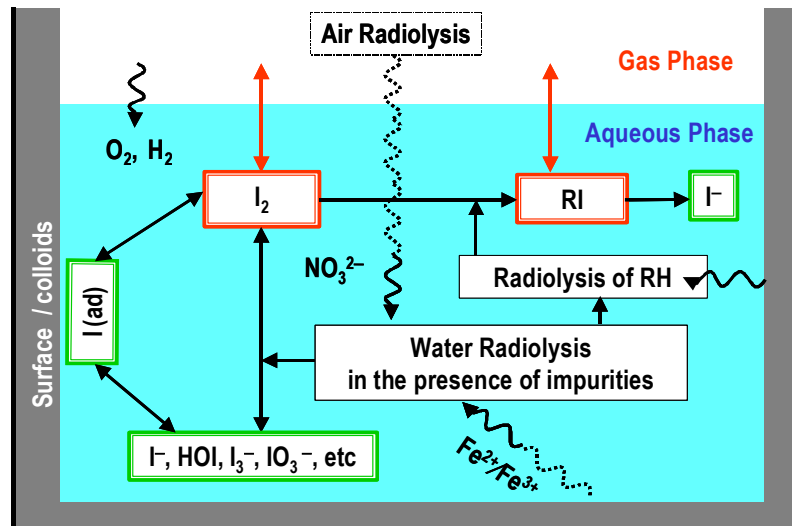


Figure 2: Schematics of Key Aqueous Processes

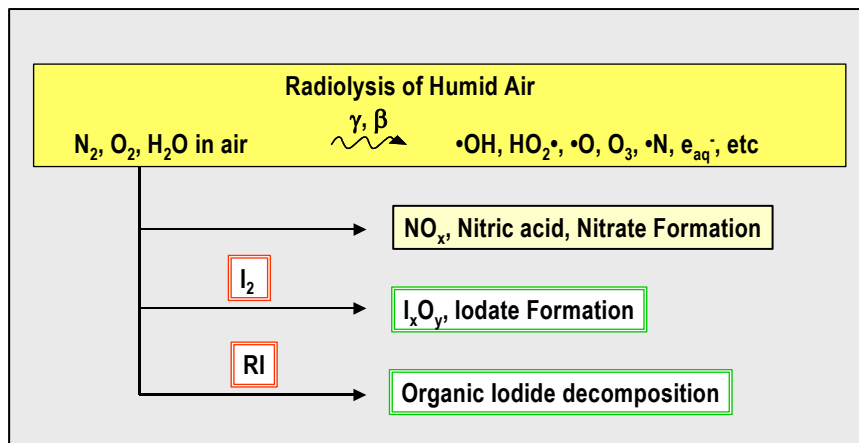


Figure 3: Key Gas Phase Processes

The transport behaviour and surface interaction of iodine is highly dependent on iodine speciation. This makes it difficult to separate the effects of individual parameters through analysis of the data collected in a limited number of engineering-scale integrated-effects tests. Consider, for example, the role of temperature in such tests. The rates of thermal reactions such as hydrolysis of I_2 or organic iodides increase with temperature, thus reducing I_2 or RI production in the aqueous phase, but the partitioning of these volatile species into the gas phase also increases with temperature. I_2 , being an oxidizing species,

will react more easily with many surfaces than I^- . Their surface interactions also have different temperature dependences. The net effect of temperature on iodine volatility is thus not straightforward and iodine volatility could either increase or decrease with temperature, dependent not only of the factors considered above but also on other conditions such as the water pH and the types of surface present.

In containment, both homogeneous (in only one phase, e.g. water) processes and heterogeneous (water-gas, gas-solid (or gas-surface), and water-solid (or water-surface)) interfacial processes occur and are inter-dependent. The homogeneous reactions are independent of the containment geometry and the makeup of the containment materials but the heterogeneous reactions could be strongly dependent on the containment design. This makes it difficult to establish a simple scaling factor when considering experimental design.

2.3 Larger Scale Tests and Models

Significant progress has been made, particularly over the last 15 years since the results from intermediate engineering-scale integrated-effects radiation test facilities, such as the Radioiodine Test Facility at AECL Canada and the CAIMAN Facility at IRSN Cadarache, France, became available. These facilities, having some control of test conditions and on-line measurements, have been used for systematic and parametric studies in multi-component environments. Integrated-effects tests carried out in these facilities have been valuable in establishing the relative importance of various chemical and transport processes and prioritizing the supporting bench-scale R&D activities that examine individual processes. They also have been instrumental in the interpretation of a much-larger scale PHEBUS-FP experiments. The integrated-effects tests performed in the intermediate-scale and large-scale facilities have been crucial for developing and validating whole system models of iodine behaviour under post-accident containment conditions.

Nevertheless, the results from the intermediate-scale test facilities only give us the net effects of many contributing phenomena or processes. Some of these processes are dependent on the geometric scale of test facility. Thus, the results from these tests are often difficult to interpret and cannot be directly applied to the real containment conditions. For the same reason, the PHEBUS results cannot be directly applied to real containment conditions. Hence, the international iodine community over the last decade or so has performed valuable focussed experiments and conducted modelling activities in support of interpreting the results of the intermediate- and large, engineering-scale studies. These research activities at more fundamental levels have made, and will make, possible the development of iodine models with better predictive capabilities.

The iodine models now available in different safety analysis codes describe iodine behaviour in varying degrees of sophistication. Mechanistic codes, such as LIRIC, INSPECT and MELCOR-I, attempt to model the whole set of radiochemical reactions and are generally used for detailed studies and interpretation of larger-scale tests. Simplified codes, such as IODE, IMPAIR, AIM and IMOD, model a reduced set of reactions and are generally incorporated in larger system-level codes. The development and continual improvement of these models over the last decade has been impressive. Particularly, over the last 7 years, the international iodine community has collaborated in the improvement of the iodine models through OECD/CSNI sponsored ISP-41 and ISP-46 code comparison exercises. The models and codes have been validated at varying degrees against separate-effect tests, intermediate-scale and integral experiments. However, these benchmark exercises also demonstrated the contribution of many user-defined model parameters or input parameters that were not well defined in the tests.

The results from the relatively recent integrated effects tests have also revealed many unexpected and synergistic effects that were not previously recognized and emphasized the complexity of underlying phenomena affecting iodine behaviour. As a result there is a need to develop strategies to identify the most important processes and questions that need to be addressed.

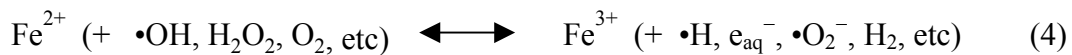
3. STEPS FORWARD

As noted above, there are a number of different ways in which additional complexity may be introduced into the modelling of iodine chemistry to better interpret large-scale tests and predict iodine behaviour in practical situations. The following sections outline areas which are judged to be particularly important and worth investigation over the next few years. This includes a brief description of the research program that is planned to be carried out at the University of Western Ontario.

3.1 The Role of Metal Ions

Water radiolysis in the presence of dissolved impurities and reactive surfaces is one of the key areas in iodine modeling which lacks quantitative understanding. The type of dissolved impurities that is of particular interest is transition metal ions. These will be present in at least trace quantities in water in contact with metal/metal oxide surfaces.

The transition metal ions are important because they have redox couples that allow the ions to participate in both oxidizing and reducing reactions. The most significant of these redox couples are $\text{Fe}^{2+}/\text{Fe}^{3+}$, $\text{Cu}^+/\text{Cu}^{2+}$, and $\text{Ni}^+/\text{Ni}^{2+}$ (because of the parent metal presence in structural materials). Through regenerative reaction cycles, these couples can allow the metal ions to participate catalytically in reactions with important species in the water radiolysis system, e.g.,



The rate constants of some of the forward reactions are reasonably well established. However, the reverse reactions are not easy to examine due to the difficulty in preparing Fe^{3+} solutions. This makes it challenging to evaluate the ratio of the steady-state concentrations of Fe^{2+} and Fe^{3+} and, hence, the overall impact of $\text{Fe}^{2+}/\text{Fe}^{3+}$ on the decomposition rates of the oxidizing and reducing species.

Different redox couples can be expected to have different impacts on the overall radiolysis chemistry because of their difference in electrochemical potential. Hence, the effect of $\text{Cu}^+/\text{Cu}^{2+}$ is expected to be greater due to lower redox potential of this couple, compared to that of $\text{Fe}^{2+}/\text{Fe}^{3+}$. A program for investigating the effects of different metal species is necessary to develop a basis for understanding the behaviour of an aqueous system with a mixture of metal ions present, and also provide insight into which metal ion species has the greatest impact. This will allow us to establish a methodology for inclusion of the metal ion reactions in our advanced radiolysis models without incurring the penalty of too much complexity. An experimental program is planned to study the impact of the redox couples of iron, nickel and copper on water radiolysis behaviour in the presence of gamma-radiation using controlled experiments in a gamma-irradiation cell and measurement of radiolysis products like H_2 . The results will be used to establish the basis for inclusion of metal ion reactions in the basic water radiolysis model.

3.2 The Role of Heterogeneous Reactions with Submerged Metal/Metal Oxide Surfaces

Submerged metal/metal oxide surfaces affect iodine chemistry through its synergistic interactions with water radiolysis products as well as with iodine species. Metal surfaces could provide catalytic sites for heterogeneous reactions such as decomposition of H_2O_2 , one of the more important stable water radiolysis products. Most of metal/metal oxide surfaces also react readily with various aqueous iodine species to form metal iodides, that may or may not be soluble in water (depending on pH, temperature, etc.). If they are soluble (e.g., FeI_2), then the reaction to form an iodide could act catalytically to transfer metal ions into solution and thereby affect the dissolution rate. If the metal iodide is insoluble (e.g., AgI) then the surface reaction could reduce iodine volatility by acting as potential iodine getter for the post-accident management. The metal/metal oxides of main interest include carbon steel and stainless steel (structural materials) and silver (either as a deliberate iodine getter or present from control rods (for some reactor designs). Understanding the influence of steel surfaces on iodine behaviour is important not only because they would be present in a containment, but also because these surfaces have been used in essentially all the large-scale iodine studies, such as in the RTF and PHEBUS FP0 and FP1 tests.

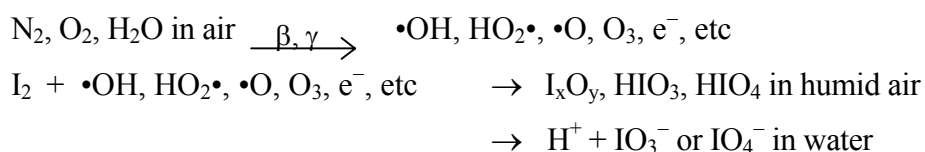
An important factor influencing the rates of reactions of aqueous iodine species is the nature of the oxide layer present on all structural metals. At the UWO an experimental program is planned to study the reaction of carbon steel, stainless steel and Zn with H_2O_2 in both the absence and presence of γ -radiation, and to then further examine the effect of that the additional presence of iodine has on the kinetics of the oxidation of metal surfaces the dissolution of the corrosion products. Different oxides with different chemical and physical properties, e.g., thickness, composition, crystallinity will be prepared thermally and electrochemically. The changes in surface properties will be studied using various electrochemical and surface analytical techniques.

3.3 The Role of Organic Surfaces

A second class of heterogeneous reactions can occur on submerged organic surfaces (such as paints). In intermediate- and large scale-tests organic surfaces have been the sources of the organic impurities in the aqueous phase and it has been difficult to unequivocally separate the role that homogeneous and heterogeneous organic reactions have played in generating the organic iodides that have been observed. Improved prediction of iodine volatility depends on understanding the dominant mechanism active in the intermediate- and large-scale tests and using this to model the dominant mechanism under post-accident containment conditions. Although there is some data available on the production of organic iodides from painted surfaces, a systematic study is warranted to better understand the competing processes. The role of organic surfaces is being addressed in various laboratories, including AECL in Canada and through EPICUR program at IRSN in France.

3.4 Effects of Humid Air Radiolysis

In the presence of radiation, gaseous I_2 could react with the radiolysis products of humid air to form solid non-volatile iodine oxides (e.g., I_2O_5 , $\text{I}_4\text{O}_9 = \text{I}_x\text{O}_y$), which would dissolve in bulk water as non-volatile iodate (IO_3^-) and periodate (IO_4^-) ions:



These processes could lower the effective iodine volatility. This conversion of gaseous iodine species by air radiolysis products is reasonably well understood and quantified in the absence of other impurities or reactive surfaces. However, analyses of various intermediate-scale tests performed in the RTF, CAIMAN and PHEBUS indicate that the level of iodine oxide formation would be much lower in containment than those observed in the laboratory-scale tests performed in clean environments.

The challenge in predicting iodine oxide formation under the post-accident containment conditions is in determining the concentrations of the air radiolysis products in the presence of reactive containment walls and aerosols. Air radiolysis is responsible for both iodine oxide formation, the decomposition of organic iodides in the gas phase and nitric acid/nitrate formation. The latter is an important process to be considered in the aqueous iodine behaviour as discussed earlier (Figure 3). Unsurprisingly nitric acid and ozone formation rates in air radiolysis have been observed to decrease considerably in the presence of reactive surfaces. However, the quantitative effect of surfaces is not well known. This deficiency will be addressed by the work of the PARIS project.

3.6 Gaseous Iodine Sorption on Surfaces

Molecular iodine, I₂, is a notoriously 'sticky' molecule with an affinity for surfaces. There are many different types of surfaces in containment, including stainless and carbon steel, aluminium, organic-based painted surfaces, bare concrete, zinc-primer coated surfaces and plastics. The temperature and relative humidity dependence of the rate of adsorption of gaseous iodine varies considerably from surface to surface. Combined with the difficulty in accurately establishing the mass transport conditions in intermediate and large-scale tests, the uncertainty in the gaseous iodine adsorption rates on containment surfaces have contributed significantly to the spread in the results observed during ISP 41 exercises.

One aspect of iodine adsorption, that on metal/metal oxide surfaces, will be investigated at UWO in collaboration with AECL. The experiments will involve in-situ measurements of adsorption under fast flow conditions using radioiodine as a tracer and non-tracer studies using the surface analyses techniques (e.g., in-situ XPS, SEM/EDX). The studies will examine the effects of reaction conditions (humidity, temperature, concentrations, etc.) and the type of metal/metal oxide (n-type and p-type) on the anion incorporation process in the oxide layer and the rate of corrosion (additional oxide formation). The experimental studies will be accompanied by chemical kinetics and mass-transport model analyses, using FACSIMILE and FEMLAB.

3.7 Integrated Effects Tests

The above list of topics for further investigation involves probing the reaction processes at fundamental levels in order to better interpret large-scale tests and extend the validity for iodine models. As our understanding of the fundamental processes improves, our ranking of the relative importance of various chemical and transport processes may change. It is thus important to maintain the integrated-effects test capability, so that we can continue to develop and validate whole system models of iodine behaviour under post-accident containment conditions. From this perspective, intermediate-scale integrated-effects test facilities, such as the CAIMAN facility, France and the ThAI facility in Germany, that can examine combinations of sub-sets of multi-component environments, but with good control of test conditions and on-line measurements for systematic and parametric studies, will be very useful.

4. CONCLUSION

We have made great strides from the early days of predicting radioiodine behaviour under accident conditions through the concerted research and development efforts of the international community. As a result, we have good basic models for use in predicting iodine chemistry under accident conditions. The focus of research can now shift to addressing the greater challenge of adding to the basic models the roles that additional chemical species and chemically reactive surfaces can play in influencing radioiodine chemistry. The development of these more sophisticated models will allow us to improve our capabilities to address more complex environments and thereby provide assurance that no major chemical process has been overlooked that could significantly invalidate the overall predictions of volatile iodine releases.

Several key additions to the current radioiodine models have been identified, including the role that metal ion redox couples can play in catalyzing reactions of water radiolysis products and the ability of metal surfaces to act as radioiodine 'sinks'. Experimental programs to address these and other areas for model improvement have been initiated. Through these programs and related studies, and the valuable results of future large-scale tests we can anticipate success in further enhancing our understanding of radioiodine chemistry.

Acknowledgement:

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