The Radioiodine Source Term and Its Potential Impact
on the Use of Potassium Iodide*

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Introduction

Since a major purpose of this work is to describe the radioiodine source term associated with light water reactor (LWR) accidents, it is perhaps appropriate to begin with its definition. In a general sense, the "source term" refers to the time-dependent escape of a particular nuclide, or of a specific isotope of that nuclide, into the biosphere in the course of a given nuclear reactor accident.

The source term can thus be considered as consisting of two components, the amount of the nuclide actually available for escape (as opposed to its inventory in the core), and the manner in which it enters the biosphere. Put another way, the source term is determined by physical and chemical processes which occur within the reactor containment and the mode of failure of the containment structure.

When viewed in this manner, it is obvious that an almost infinite number of possibilities exist. Moreover, even if we were to group these possibilities in some logical way, as was done, for example, in the Reactor Safety Study,[1] the problem of providing quantitative descriptions of the radioiodine source term still remains formidable. For this reason, we shall focus the presentation on more qualitative aspects.

Chemical Forms of Fission Product Iodine in the Primary Circuit

Fission product iodine is generally formed within the uranium dioxide fuel crystal lattice as a highly energized atomic species. It remains there as an impurity atom, until thermal motion causes its transport to a crystal imperfection, such as a grain boundary, or to the interconnected void region within the fuel rod. In these locations, the highly reactive atomic species comes into contact with other chemical

components within the fuel rod, so that the opportunity for chemical reaction arises.

In fact, there is now little doubt that the predominant form of fission product iodine that is contained within the interconnected void region, the so-called "gap inventory," is not highly volatile elemental iodine, but rather a chemically combined form.[2] Thermodynamics calculations[3] and indirect experimental evidence[4,5] suggest that the chemical compound is cesium iodide.

It is also likely that the iodine contained along grain boundaries or other crystal imperfections exists as cesium iodide. However, the precise mechanism for the formation of cesium iodide, and particularly the rate of formation, remains to be defined. The rate of formation is especially important, as it may be the principal determinant of the chemical form of iodine which escapes from failed fuel rods at very high temperatures, in excess of 1400°C (2550°F). At these temperatures, release from the fuel is believed to occur directly from transport through the crystal lattice, with no opportunity for reaction at grain boundaries.

In contrast to severe core damage accidents, during which fuel and cladding temperatures can greatly exceed 1400°C (2550°F), successfully terminated loss-of-coolant accidents do not involve cladding temperatures in excess of 1200°C (2200°F). As a consequence, only the gap inventories are involved in the release from ruptured fuel rods and, as has already been indicated, this implies that the release of fission product iodine is predominantly as cesium iodide. Moreover, because only the gap inventories are involved, successfully terminated loss-of-coolant accidents

are expected to result in the release of about 1% of the inventories of the long-lived noble gases, and about 0.05% of the long-lived iodine isotopes from the core of the reactor.[6] In addition, since the escaping fission products have ample opportunity to be contacted by water, and since no threat to the integrity of the containment is involved, these accidents may be regarded as mere nuisances, even though a costly cleanup operation may result.

As fuel cladding temperatures rise above 1200°C (2200°F), considerable hydrogen is produced from the reaction of the Zircaloy cladding with the steam. As a consequence, one can state categorically that all LWR loss-of-coolant accidents occur under conditions in which the environment in the primary circuit is chemically reducing. (There is one class of exceptions; these are accidents that involve catastrophic failure of the reactor vessel, and are not considered further in this work.)

Under the proper set of conditions of temperature and concentration, it does appear possible to decompose cesium iodide in a steam-hydrogen atmosphere,[7] but the decomposition products would reform the iodide as the mixture is cooled during its transit from the core to the breach in the primary circuit which had caused the loss of coolant. Or, if the cesium reaction product (which is actually cesium hydroxide) were to be preferentially removed from the mixture, hydrogen iodide could be formed. In any case, because of the chemical reactivity of elemental iodine and of the chemically reducing conditions that prevail within the primary circuit during a loss-of-coolant accident, there is little doubt that the reduced form of iodine, that is, an iodide, will emerge from the primary circuit. Moreover, this contention is supported by chemical analysis of

the water in the Three Mile Island Unit 2 (TMI-2) reactor containment building.[8]

If the temperature of a reactor core were to rise uniformly in the course of a loss-of-coolant accident, there would be a progression in the releases of the fission products and other core materials in accordance with the volatilities of the corresponding chemical species. As is evident from an examination of the TMI-2 "Quick Look" video tapes, however, rather exreme gradients in temperature, both axially and radially in the core, actually occur. Consequently, while some regions of the core may be relatively cool, so that only more volatile fission products are being released from the fuel, other regions may be sufficiently hot that even control rod and structural materials are being vaporized. This could lead to the production of aerosols, and the subsequent attachment of iodine-containing species to these aerosols. Thus fission product iodine could emerge from the primary circuit and enter the containment building in a variety of physico-chemical forms, but predominantly in the chemically reduced state.

Chemical Forms of Fission Product Iodine in the Containment Building

For purposes of this work, the containment building is defined to include all structures external to the primary circuit which are designed to prevent the escape of fission products into the biosphere. Thus, the wet well and the dry well of a boiling water reactor are considered to be part of its containment building.

As the fission product iodine emerges from the primary circuit, the chemical environment changes from a reducing atmosphere to an oxidizing

atmosphere. Because the iodine is in a chemically reduced state, oxidation is possible at this stage. And one of the possible oxidized forms is elemental iodine.

Before proceeding further, it is instructive to consider the manner in which the fission product is introduced to this change in chemical environment. Two modes are possible. In one, the fission product release pathway is intercepted by water, whereas the second involves introduction of the fission product directly into the containment building. This second mode is the so-called "dry accident." It should be kept in mind, however, that for the dry accident to occur, it is first necessary to expel some tens of thousands of gallons of water into the containment building, along the same pathway as that taken by the fission products. So even in a dry accident, significant quantities of water are nonetheless present.

If the release pathway is intercepted by water, a large reduction of the gas-borne fraction of fission product iodine is expected to occur, regardless of its physico-chemical form. This is due to the efficient manner in which aerosols are scavenged under the conditions that are expected to prevail, and the high solubilities of most of the pertinent iodides. And although elemental iodine is itself considerably less soluble than the iodides in question, it reacts chemically with water to yield nonvolatile forms. So even if elemental iodine were involved, attenuation of the airborne concentration would result.

Whether or not chemical changes in the iodine species will, in fact, occur in a dry accident will depend on the temperature at which the reduced form is introduced into the oxidizing environment. If the

reduced form is cesium iodide, then no change is expected, unless the temperature of the system is above about 700°C (1290°F);[2] this would probably require the combustion of hydrogen in the containment building. If the reduced form is hydrogen iodide, decomposition could be effected at considerably lower temperatures.

In either case, decomposition would give rise to the formation of elemental iodine. This form would then persist until it reacted with water, recombined to form cesium iodide as the system temperature is lowered, reacted with other materials present in the containment building, or escaped into the biosphere.

Oxidation of the dissolved iodide ion can also occur in the wet accident, but the process is very slow under the conditions of interest,[9] and the elemental iodine that would result, would itself react with the water. Radiation can accelerate the oxidation process, but it apparently accelerates further oxidation of the elemental iodine as well, to form the nonvolatile iodate ion.[10]

Finally, it is necessary to consider the question concerning the appearance of volatile organic iodide compounds. The precise mechanism for the formation of these species remains undetermined, but the organic iodides can be the dominant form of gas-borne fission product iodine in a reactor accident. The amounts involved are quite small, however, so they become the dominant form by default, that is, not because they are present in considerable quantity, but because other volatile forms of iodine are not present.[2]

Summary of Iodine Chemistry in Light Water Reactor Accidents

Because of the chemically reducing conditions that are character-
istic of all light water reactor loss-of-coolant accidents, the chemi-
cal form of fission product iodine that emerges from the primary circuit
and enters the containment building is a chemically combined form, an
iodide, rather than elemental iodine. Moreover, unless the cesium that
simultaneously escapes from the fuel in about a tenfold excess is somehow
separated from the iodine, the iodide compound is most likely cesium
iodide. This compound can condense on cooler surfaces of the primary
circuit, perhaps to be re-evaporated later on in the accident, or it may
deposit on aerosol particles which may be present, or it may form aerosol
particles itself.

If the iodide species is contacted by water in the course of its
release into the containment building, as had occurred at TMI-2 and as it
is designed to do in boiling water reactor systems, significant dissolu-
tion or entrapment of the iodide is expected. In this event, the
radioiodine source term will be very small and will be dominated by the
formation and escape of organic iodide compounds.

.On the other hand, if the iodide is injected directly into the con-
tainment building, its escape will be determined by its behavior as an
aerosol. However, it is important to note that the aerosol particles may
be cesium iodide crystals or may involve cesium iodide or other iodides
which are deposited on aerosol particles that are radiotoxic in their own
right.

Finally, in the event the stream of fission product iodine emerging
from the primary circuit, or parts of the containment building, are at

elevated temperatures (as the result of hydrogen combustion, for example),
then oxidation of the iodide can occur. This can lead to the production
of elemental iodine. The source term will then be determined by the rate
of recombination of the iodine with cesium, to form cesium iodide, by the
extent of subsequent contact by water, by the extent of reaction with
other materials in the containment building, and by the degree to which
the species becomes sorbed on aerosol particles.

Impact of the Radioiodine Source Term on the Potassium Iodide Issue

The issue regarding radioiodine releases in those accidents in which
substantial amounts of water are present, and in which reasonable con-
tainment integrity is maintained, appears to be resolved. The original
contention,[11] that the risk to the general public presented by iodine
in these cases is lower than previously estimated, does not appear to be
at issue. The argument concerning the distribution of potassium iodide,
KI, thus pertains to a consideration of those accidents in which large
amounts of water are not involved, and containment integrity is severely
compromised.

For this class of accidents, the current state of knowledge is in-
adequate to permit a resolution of the KI issue based upon source term
considerations alone. Nor is it necessary to base the resolution sole-
ly on source term considerations.

Three other aspects which have bearing in this regard and which will
be discussed here include: (1) the period of time available from the
indication of the likelihood of significant radioiodine release to its
actual occurrence; (2) the expected frequency of occurrence of reactor

accidents in which the risk to the general public from radioiodine might merit KI distribution; and (3) the effectiveness of KI compared with other alternatives.

Regarding the first of these, the Reactor Safety Study estimates warning times for evacuation to range beween one and two hours.[1] There is at present little reason to alter these estimates based solely on current information concerning iodine fission product chemistry. Moreover, in past accidents (TMI-2 and Windscale being the most notable), the first realization of the seriousness of the situation had come only with the detection of large releases of radioactivity. On the other hand, such releases should not be taken as a signal for the deployment of thyroid blocking agents. As occurred at TMI-2, although some 8 million curies of radioxenon were released into the biosphere, at no time was the public at risk due to radioiodine exposure.

Accidents which can potentially involve the escape of large quantities of radioiodine are those in which the release pathway avoids contact by water, and early containment failure occurs. These accidents are estimated to happen with frequencies in the range of 10^{-5} to 10^{-7} per reactor year.[1] With about 100 reactors operating in this country, it is thus pertinent to inquire about the logic of maintaining a continuous personal stock of KI doses to contend with an event which will occur at a rate of perhaps once every ten centuries. The number of fatalities which is projected to occur over this period, due solely to accidents associated with the distribution of the drug, is certainly not insignificant, even if a 10-year shelf-life of the drug were assumed.

Lastly, it has been estimated that the administration of 100 mg doses of KI to adults could result in a tenfold reduction in the incidence of typhoid cancer in the event of exposure to radioiodine.[12] However, administration of the drug is itself not without risk; this was in fact recognized in the study of the American Physical Society.[12] The magnitude of the risk involved remains a subject of intense discussion.

In addition, studies of the source term suggest little likelihood of significant releases of elemental iodine, that is, chemically uncombined iodine, in severe core damage accidents. The studies indicate release of cesium iodide as particulates or as attached to aerosol particles which may themselves contain radiotoxic species. Moreover, even elemental iodine would tend to be associated with particulates. The use of KI, which is effective only for radioiodine exposure of the thyroid, may thus address a relatively minor part of the issue and may, in fact, exacerbate the consequences of a reactor accident if the administration of KI were to render a false sense of protection to an uninformed user.

The use of KI, even as an interim measure until the many questions associated with severe accident consequences are delineated, remains an intense issue. The question then arises as to whether or not a more acceptable alternate to KI is available. One such alternate is suggested by the effectiveness of water in reducing the concentration of iodine in reactor accidents, viz., the use of moistened cloth, such as a handkerchief.

Studies of the effectiveness of moist cloth indicate a two-fold reduction in elemental iodine concentration by six layers of moist cloth, and a two- to ten-fold reduction in aerosol concentration, depending upon

aerosol particle size.[13] Moreover, the tests with iodine were conducted

at iodine concentrations which were several orders of magnitude larger

than would result in a severe core damage accident. More recent data,

taken at significantly lower concentrations, indicate that a factor of

ten reduction in iodine concentration is readily attainable.[14] The use

of such material can thus afford protection not only from the effects of

radioiodine inhalation, but from other radiotoxic species as well.

This proposed alternative to the distribution of KI is of course not

new. Nor is it entirely free from objection. Nonetheless it, and other

alternatives which have also been proposed, should be joined to the issue.

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