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Original Article

RAIM – A MODEL FOR IODINE BEHAVIOR IN CONTAINMENT UNDER SEVERE ACCIDENT CONDITION

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ABSTRACT

Following a severe accident in a nuclear power plant, iodine is a major contributor to the potential health risks for the public. Because the amount of iodine released largely depends on its volatility, iodine's behavior in containment has been extensively studied in international programs such as International Source Term Programme-Experimental Program on Iodine Chemistry under Radiation (EPICUR), Organization for Economic Co-operation and Development (OECD)-Behaviour of Iodine Project, and OECD-Source Term Evaluation and Mitigation. Korea Institute of Nuclear Safety (KINS) has joined these programs and is developing a simplified, stand-alone iodine chemistry model, RAIM (Radio-Active Iodine chemistry Model), based on the IMOD methodology and other previous studies. This model deals with chemical reactions associated with the formation and destruction of iodine species and surface reactions in the containment atmosphere and the sump in a simple manner. RAIM was applied to a simulation of four EPICUR tests and one Radioiodine Test Facility test, which were carried out in aqueous or gaseous phases. After analysis, the results show a trend of underestimation of organic and molecular iodine for the gas-phase experiments, the opposite of that for the aqueous-phase ones, whereas the total amount of volatile iodine species agrees well between the experiment and the analysis result.

1. Introduction

Following a severe accident in a nuclear power plant, iodine is a major contributor to the potential health risks for the public, especially with regard to the thyroid. During a core meltdown, a large fraction of iodine in the core can be released into the containment in aerosols form, containing metal iodides and gaseous iodine, as found in Phébus tests [1–3]. Gaseous iodine released from the reactor coolant system (RCS) is depleted mainly through trapping on the containment surface, which can result in the production of organic iodide through interactions between iodine and paint [4–6]. The gaseous molecular iodine could also react with air radiolysis products (ARPs) such as ozone, to form iodine oxide; an aerosol-borne iodine species [7,8]. Organic iodides are also depleted by radiolytic decomposition. However, most metal-iodide

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particulates can be readily dissolved in sump water resulting in iodide ions. These will be oxidized to form volatile inorganic iodide (I_2) through many reactions such as radiolysis and hydrolysis. The organic radicals, made from organics such as paint in the sump water, react with the molecular iodine to produce organic iodides. These gaseous iodine species move from the sump water to the containment atmosphere mainly by diffusion and natural convection [9].

The amount of iodine that could be released from the containment largely depends on the volatility of the various kinds of its species [1] and their retention by filters used in the containment venting systems. Therefore, iodine behavior has been a major topic of international research programs such as International Source Term Programme (ISTP) [7,8,10], Organization for Economic Co-operation and Development-Behaviour of Iodine Project (OECD-BIP) [11] and Organization for Economic Co-operation and Development-Source Term Evaluation and Mitigation (OECD-STEM) [12]. The major research areas of these programs are iodine chemistry, surface reactions, mass transfer, modeling of iodine chemistry and its applications to severe accident assessment, and accident management. Qualified tools for modeling these phenomena have been developed and validated experimentally.

ISTP was carried out by the IRSN (Institut de Radioprotection et de Sûreté Nucléaire) for 5 years until the end of 2010 in order to reduce uncertainties when evaluating the environmental release of radioactive products such as iodine or ruthenium following a core meltdown accident in a pressurized water reactor (PWR). Among its various experimental topics, EPICUR and PARIS programs provided experimental data on the physicochemical transformations of iodine (formation and destruction of volatile iodine species) under the effect of radiation in the reactor containment. OECD-BIP was organized by the Organization for Economic Co-operation and Development, Nuclear Energy Agency (OECD/NEA) and has been operated by Canadian Nuclear Laboratories (former AECL) since 2008, performing work in the following three areas: adsorption and desorption of iodine on surfaces, organic iodide formation from containment paints loaded with iodine from the gas or aqueous phases, and provision of Radioiodine Test Facility (RTF) experimental data from historical experiments. OECD-STEM was launched in 2011 to better address known phenomenological uncertainties relevant to radioactive iodine release in the mid and long term time frame, especially in terms of the stability of iodine aerosol particles under radiation, the long-term gas/deposits equilibrium in a containment, and iodine interactions with paints. Ruthenium chemistry was also investigated in this study.

The Korea Institute of Nuclear Safety (KINS) has been studying the iodine chemistry issue as one of the regulatory research topics, and has joined international programs with other Korean organizations such as the Korea Atomic Energy Research Institute (KAERI). In the course of this study, a simplified, stand-alone iodine chemistry model, RAIM (Radio-Active Iodine chemistry Model) has been developed in order to deal practically with iodine chemistry in the containment [13,14]. This model is based on the semiempirical IMOD methodology [15,16] in terms of the simplification of the chemical species, reaction mechanisms, (especially for the aqueous phase), and reaction rate constants. The gas-phase models are largely based on other previous studies carried out by Bosland et al [7,17] and Funke [18]. Although RAIM is still under development, attempts have been made to couple it with the MELCOR code [19] in order to model organic iodides and to simulate Phébus FPT1 and FPT3 tests [20]. However, the MELCOR-RAIM estimation of the organic iodide concentration was very small compared to the measured data, which showed the need for the further development of its homogeneous reaction model of organic iodide formation. This article summarizes the activities concerning the development of the RAIM model and the benchmarks of the ISTP-EPICUR S1 and S2 series and OECD-BIP P10T2 experiments.

2. Model development

In order to develop an iodine chemistry model, the existing methodologies have been reviewed. There are two types of iodine behavior models: mechanistic and semiempirical models. Mechanistic models such as LIRIC [21,22], INSPECT [23], and MELCOR [19] are based on detailed data established through extensive analysis of iodine reactions; they also include mechanisms and intermediate products of iodine reactions. However, empirical models such as IMOD have an advantage with regard to their compatibility with integrated severe accident codes due to their simplified scheme, even though they may have limitations in terms of providing accurate calculations for such complicated iodine reactions [9,13].

Following previous studies, we decided to develop our own model based on the LIRIC methodology [20,21], but aiming for a smaller and simpler model similar to IMOD [15,16]. Therefore, reactions in the aqueous phase and volatile species for mass transfer were reduced. For the simplification of reactions and rates, single species were used in the gas and aqueous phases. For instance, the iodine species to be examined were volatile I_2, organic iodides of high volatility (HVRI) and low volatility (LVRI), nonvolatiles, nonaqueous iodine, iodine oxide aerosols (IO_x), and iodine adsorbed on the surface (I_{ad}) in the gaseous phase [15,16]. HVRI refers to all of the organic iodides that are volatile, e.g., CH_3I which is more volatile than I_2, whereas LVRI includes all of the organic iodides that are less volatile. Nonvolatile iodine species in the aqueous phase consists of mainly iodide ion, $I^-_{(aq)}$, with minor components of species such as HOI, I_3^-, and IO_x^- including IO_3^-. Nonaqueous iodine species are those bound on the surfaces in contact with the aqueous phase, e.g., AgI colloids and solids, or iodine adsorbed on surfaces in contact with the aqueous phase [16]. Several other materials participating in the iodine reactions, e.g., organics and organic radicals, and ARPs such as ozone, are also dealt with. Fig. 1 shows the reactions and the transport processes of the iodine species that are modeled by RAIM.

The reaction rate constants suggested by previous studies [8,15–18,21,22] have been adopted; some data from them were given as functions of temperature or the associated activation energy. Reactions with no available rate constants were determined through simulation and sensitivity analysis of EPICUR and RTF experiments. The differential equations for the kinetics of the reaction schemes are solved numerically