Observations Emerged from ISTP-EPICUR on Iodine Behavior During a Severe Accident

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1. Introduction

Iodine behavior is most important in the evaluation of the early effects of a severe accident on the off-site public health. However, the MELCOR code, which is being used by Korea Institute of Nuclear Safety (KINS) for an integrated severe-accident assessment, does not treat the organic iodine behavior in the containment atmosphere. Therefore, it is needed to develop a stand-alone model which can be coupled with the current radiological assessment methodology.

KINS agreed with other domestic organizations, including Korea Atomic Energy Research Institute (KAERI), Korea Hydro & Nuclear Power Co. Ltd. (KHNP) and KEPCO Engineering & Construction Company, Inc. (KEPCO E&C), to participate in the ISTP (International Source Term Program) operated by IRSN. This Program investigates the behaviour of iodine and ruthenium in the containment of a power plant in case of severe accident. It consists of two sub programs: ISTP-EPICUR and PARIS. This study was carried out to evaluate the experimental results and derive observations that could be used for development of an iodine behavior model and its validation [1].

2. Experimental Program of ISTP

2.1 EPICUR Experiment

IRSN has performed the EPICUR tests which consist of experiments concerning mass transfer, radiolysis in the sump, organic iodide formation from the painted coupons, stability of iodine oxides and aerosols, and a qualification experiment. It consists of four test series; for the S1 test series painted coupons were put in the iodine solution whereas iodine-loaded coupons were put in the gas phase for the S2 series. The S3 series had an objective to examine the destruction of iodine oxides and S4 series was carried out to study volatilization of iodine from CsI aerosol deposited on the coupon.

The EPICUR loop allows continuous γ measurement of the rate of gaseous iodine volatilization under radiation. As a schematic view of the loop is shown in Fig. 1, the irradiation vessel is connected through stainless steel tubes to the May-pack device. The May-pack system, which consists of several stages equipped with quarts fiber filter, Knit-mesh and impregnated charcoal filters, can retain iodine aerosols, molecular iodine and organic iodides each. The volatile species produced in the irradiation vessel are transferred to the May-pack device with the gas flow bubbling through the gaseous phase. On-line γ measurements are provided by NaI(Tl) counters placed above each stage of filters of the May-pack device [2].

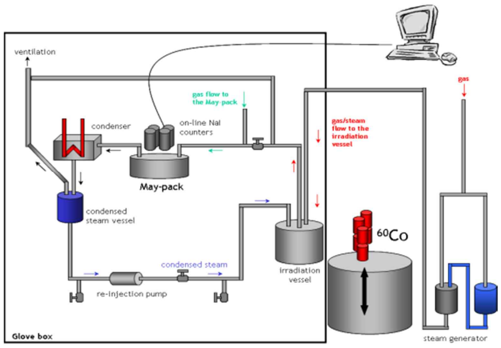

Fig. 1. Simplified view of the experimental EPICUR loop.

2.2 Ruthenium Experiment

Ruthenium is an element that has chemical toxicity and high radioactivity as well. It could occupy 15% to 20% of the total activity of all the fission products during a severe accident. As a part of ISTP, experimental study of stability and adsorption of the volatile gas, RuO_4(g), on specific surfaces in the containment were carried out (See Fig. 2). Analytical experiments on the ruthenium chemistry such as production of RuO_4(g) from the oxidation of RuO_2, and from the ruthenium precipitates adsorbed on the painted coupon, and from ruthenium aerosols and RuO_4(aq) in the containment sump have been performed [3].

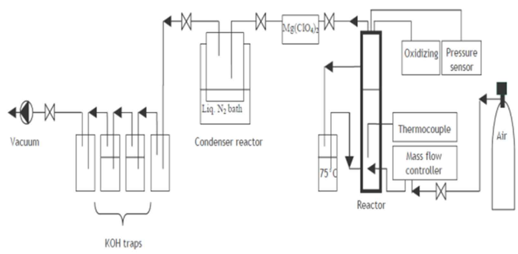

Fig. 2. Scheme of the experimental set-up for generating and trapping crystalline RuO_4.

2.3 PARIS Experiment

The PARIS experimental program (2003 ~ 2005) was composed of eleven irradiation tests to observe the effects of air radiolysis and surfaces on the gaseous iodine. Irradiation tests were conducted with Co-60 source which provided dose rates of several kGy/h and under the temperature of 80°C or 130°C. This program was extended to the EPICUR experiments [4].

3. Observations emerged from ISTP

3.1 EPICUR Experiment

Fig. 3 shows a typical online activity measurements at the quarts fiber filter, the Knit-mesh and the impregnated charcoal filters in the EPICUR experiment. The transfer rates of molecular iodine and organic iodide followed a logarithmic law [2].

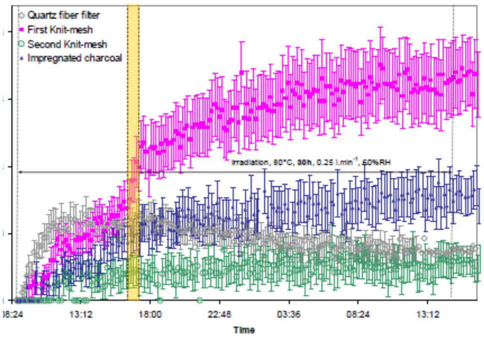

Fig. 3. Online measurements of the quarts fiber filter, the Knit-mesh filters and the impregnated charcoal filters.

Effects of various test parameters on the volatility of iodine were shown in the tests. The volatilization rate for molecular iodine estimated from the online measurements at 80°C, pH of 7 was approximately two orders of magnitude lower than the molecular iodine transfer rates determined under the similar conditions at pH 5. This seemed to be consistent with the NUREG-1465 accident source term [5]. Total quantity of the volatilized iodine was higher at 80°C than at 120°C, and was higher with the relative humidity of 60% than with 0%, independently of the volatile species. Recent tests such as S2-6 series showed that the more iodine were loaded on the coupon, the less fraction of iodine were volatilized in the form of organic iodide; this trend complies with that of OECD-BIP (RTF experiment). Long irradiation of the painted coupon consistently made slow volatilization of iodine even after 8 hours [2].

3.2 Ruthenium Experiment

Ruthenium tetroxide (RuO_4(g)) was not so stable under severe accident conditions but its decomposition rate was not as fast as expected. The product of decomposition was mainly the ruthenium dioxide. During the irradiation test, increase of the temperature and the humidity clearly enhanced the volatilization of ruthenium precipitates adsorbed onto the painted coupon. Irradiation tests in aqueous phase showed that there was no ruthenium volatilization from RuO_2 aerosol. However, the volatilization became significant in the case of RuO_4^- species in an acid medium, and even more fraction of ruthenium volatilization occurred when the gaseous forms of ruthenium tetroxide (RuO_4(aq)) are transferred into either the basic or acidic sump [3].

3.3 PARIS Experiment

Main observations from the PARIS experiment were as follows; the concentration of air radiolysis products (ARP) generally increased with dose. Silver surfaces slightly decrease ARP concentrations. However, other conditions such as temperature, dose rate, hydrogen, air or oxygen, stainless steel surfaces or painted surfaces did not affect ARP formation significantly.

In the absence of surfaces, I_2 was effectively converted by radiation into iodine oxides. Formation of IOx increased with decrease of I_2 concentration and temperature. Surfaces acted as traps for iodine, as expected. Silver, with clearly the highest S/V ratio, was the most efficient trap for iodine. Paint showed relatively smaller iodine loadings and the lowest iodine loading was observed on stainless steel. Deposition on surfaces was stronger at longer exposure times [4].

4. Conclusions

In the EPICUR experiment, iodine species were measured through the online facility and the post-test gamma spectrometry. Effects of various parameters on the iodine behavior were examined. Similar study was carried out for the volatilization of ruthenium. In the PARIS experiments, the effects of air radiolysis and surfaces on the gaseous iodine were observed. The experimental data, together with those of OECD-BIP will be used for development of the evaluation model for iodine behavior in the containment. In addition this information could be also used for development of the domestic accident source term for safety analysis.

Acknowledgments

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