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ATR-2 Part B: Ruthenium transport in an RCS with airborne CsI

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Abstract

In the NKS-R ATR-2 activity (year 2015) by VTT Technical Research Centre of Finland Ltd and Chalmers University of Technology the aim was to study the effect of the air radiolysis products N_2O, NO_2, HNO_3 (see Part A) and CsI aerosol (see Part B) on the transport of gaseous and particulate ruthenium species through a model primary circuit. The main outcomes of the airborne CsI effect on Ru behaviour are shortly summarized in this document (Part B). A detailed description was published as a scientific journal paper (open-access):

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Key words

Ruthenium, Caesium Iodide, Severe Accident, Source Term

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Research Report of the NKS-R ATR-2 activity

(Contract: AFT/NKS-R(15)111/2)

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Summary

In the NKS-R ATR-2 activity (year 2015) by VTT Technical Research Centre of Finland Ltd and Chalmers University of Technology the aim was to study the effect of the air radiolysis products N_2O, NO_2, HNO_3 (see Part A) and CsI aerosol (see Part B) on the transport of gaseous and particulate ruthenium species through a model primary circuit. The main outcomes of the airborne CsI effect on Ru behavior are shortly summarized in this document (Part B). A detailed description was published as a scientific journal paper (open-access):

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Preface

The experiments on the transport of Ru in primary circuit conditions were conducted at VTT Technical Research Centre of Finland Ltd (Espoo, Finland) in 2015. The "VTT's Ru transport facility" was slightly updated for these new experiments. The samples of gaseous and particulate ruthenium, produced as a result of experiments, were analysed with various techniques at Chalmers University of Technology (Göteborg, Sweden) and VTT (Espoo, Finland). The activation of Ru samples for INAA analysis was performed with VTT´s research reactor.

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Authors

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Summary of the experimental study on Ruthenium transport in an RCS with airborne CsI

The main outcomes of the airborne CsI effect on Ru behavior in a reactor coolant system (RCS) are shortly summarized in this document (Part B of the NKS-R ATR-2 activity). A detailed description was published as a scientific journal paper:

Kärkelä, T., Kajan, I., Tapper, U., Auvinen, A., Ekberg, C. 2017. Ruthenium transport in an RCS with airborne CsI, Published online 5th of May, 2017 (Progress in Nuclear Energy), http://dx.doi.org/10.1016/j.pnucene.2017.04.019 (available as an open-access publication).

Summary (referring to [1])

Release of the fission product ruthenium from damaged nuclear fuel occurs when the metallic ruthenium is oxidized to gaseous RuO_2, RuO_3 and RuO_4. When the temperature decreases below approx. 1000 K, the released RuO_2 has already condensed to solid RuO_2, whereas RuO_3 has decomposed to RuO_2 and then also condensed to solid RuO_2. Therefore, only RuO_4 can be observed in gaseous form at low temperatures. The impact of oxidizing conditions on ruthenium release and transport has been studied previously. The main emphasis has been on ruthenium chemistry in pure air and steam/air atmospheres. In large-scale Phébus FP experiments [2, 3] it was observed that most of the released ruthenium was transported to the containment building as solid RuO_2. Small-scale experiments [4 - 6] have shown that the transport of gaseous ruthenium through a reactor coolant system into the containment building can be much higher than would be expected on the basis of thermodynamic equilibrium calculations. It was observed that the decomposition of gaseous RuO_4 was not complete and it did not follow the equilibrium model when the residence time of gas flow was short in the area of high temperature gradient in a model primary circuit [7]. As a result, the observed partial pressure of RuO_4 in containment conditions was on a level of 10^{-6} to 10^{-8} bar at 310 K to 400 K. The research on air ingress conditions was taken even further in a recent study [8] (Part A of the NKS-R ATR-2 activity), in which the air radiolysis products N_2O, NO_2 and HNO_3 with representative concentrations were fed into the flow of ruthenium oxides in a model primary circuit. Both NO_2 and HNO_3 appeared to be efficient in oxidizing lower ruthenium oxides to RuO_4 and increasing the transport of gaseous ruthenium beyond the previous observations in pure air and steam/air atmospheres. [1]

In addition to gaseous additives, the gas flow in a reactor coolant system in severe accident conditions also includes aerosols, such as fission products and control rod materials. The effect of aerosols on the transport and speciation of ruthenium has not been studied extensively. In a previous study with silver particles fed into the flow of ruthenium oxides [9] (as part of the NKS-R ATR-1 activity), the transport of gaseous ruthenium decreased significantly as RuO_4 condensed to RuO_2 on the surface of silver particles. Another representative compound in the primary circuit is radiotoxic caesium iodide, which is the most important form of iodine transported into the containment atmosphere [10]. There is some evidence of the retention of ruthenium on a surface coated with caesium at high temperature (750 to 900 K) [11] and of the trapping of gaseous ruthenium by CsI deposit at low temperature (ca. 300 K) [5]. However, the effect of airborne CsI on the transport of ruthenium in an RCS is not known. The aim of this study was to focus on the behaviour of Ru-CsI system in the gas phase and to determine experimentally whether CsI would be able to affect the ruthenium transport through an RCS in air ingress conditions. [1]

All the experiments were conducted with VTT´s Ru transport facility. The RuO_2 precursor was heated inside a furnace up to 1300 K, 1500 K and 1700 K under dry or slightly humid air atmosphere and the formation of gaseous ruthenium oxides took place. Additional CsI aerosol was fed into the flow of ruthenium oxides inside the furnace. The airborne CsI had a significant effect on the thermodynamic equilibrium of Ru species. In comparison to pure air atmosphere, the transport of gaseous ruthenium increased from 0.2 % up to 16 % and 6 % at 1500 K and 1700 K, respectively, whereas the aerosol transport of ruthenium decreased significantly. Thus, the gaseous Ru transport corresponds to a partial pressure of 10^{-5} bar. This is the highest amount of Ru ever observed in gaseous form in the experiments with this facility. At 1300 K the transport of ruthenium was rather similar to air atmosphere and the partial pressure of gaseous ruthenium was 10^{-6} bar as it has been reported previously.

The properties of the formed ruthenium containing aerosol particles were online monitored with several measurement devices. The transport of aerosol particles was proceeding with a constant rate in the experiments. The most of the particles were approximately 100 nm in diameter, although a significant fraction of the particle mass was observed for particles with a diameter above 1 µm. Based on the XPS analysis, the transported ruthenium containing aerosol was of RuO_2. The shape and size of the crystals changed when CsI was mixed with Ru oxides in the gas phase inside the furnace. The obtained aerosol measurement data (with two devices utilizing different operation principles) was used to derive the shape factor for the RuO_2 aerosol. The shape factor plays a role when considering the aerosol transport, therefore the shape factor is required especially in severe accident computer models to give reliable predictions on aerosol behaviour.

These obtained results indicate that the airborne CsI can effect on the fraction of gaseous ruthenium reaching the containment building during a severe nuclear accident in case of air ingress into the reactor. The formation of gaseous ruthenium in significantly higher extent than what has previously been expected needs to be considered also in the current severe accident computer models, which are currently lacking of this information. Given the potential radiological consequences of radiotoxic ruthenium to the population in case of a source term, and the high priority research topic label issued by the former SARNET-2 network, the effect of CsI and other fission products, structural materials etc. on the ruthenium chemistry need to be investigated in detail.

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