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Transport of ruthenium in primary circuit conditions during a severe NPP accident

T. Kärkelä [a,*], N. Vér [b], T. Haste [c], N. Davidovich [d], J. Pyykönen [a], L. Cantrel [c]

[a] VTT Technical Research Centre of Finland, P.O. Box 1000, FI-02044 VTT, Finland
[b] Centre for Energy Research (EK), Hungarian Academy of Sciences (MTA), P.O. Box 49, 1525 Budapest 114, Hungary
[c] Institut de Radioprotection et de Sûreté Nucléaire (IRSN), PSN/SAG/LETR, CE Cadarache, F-13115 Saint Paul lez Durance, France
[d] Italian National Agency for New Technologies, Energy and Sustainable Economic Development (ENEA), C.R. Casaccia, SP 101 via Anguillarese 301, 00123 Roma, Italy

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ABSTRACT

Ruthenium species, volatilized from damaged fuel during a severe accident in a nuclear power plant, are radiotoxic and can be transported to the containment atmosphere in gaseous form. To limit the possible source term to the environment, it is of interest to understand the behaviour of Ru after it has been released from fuel and the phenomena taking place within the decreasing temperature section of the reactor coolant system. This was investigated in the framework of EC SARNET and SARNET2 projects, as a part of the Source Term work package, with several separate-effect tests on the transport and speciation of Ru in primary circuit conditions considering the influence of other fission products as well. The source of Ru was metallic Ru, RuO_2 powder or gaseous RuO_4. The large-scale integral tests of the Phébus FP program were conducted with real irradiated fuel, and more realistic analysis on the release and transport of Ru could be performed. Experimental studies proved that the transport of ruthenium to the containment atmosphere took mainly place as RuO_2 particles when Ru source was oxidized above 1250 °C. The fraction of Ru transported in gaseous form was at its highest when ruthenium was oxidized at approx. 1000–1100 °C. A major part of the released Ru was deposited at the decreasing temperature area of the circuit as RuO_2. Revaporisation of the deposited Ru at low temperature was a significant source of gaseous ruthenium. In order to understand the behaviour of ruthenium in these tests, the analysis work was extensive and several simulations were carried out. As an outcome, the observed transport and deposition of ruthenium was explained. The simulation studies gave also an insight into the performance of the ASTEC code and some model improvements for Ru transport through the reactor coolant system have been identified.

1. Introduction

The effect of oxidizing atmosphere on source term has been one of the high priority open issues in the framework of EC SARNET and SARNET2 (Severe Accident Research NETwork of Excellence) projects (Schwinges et al., 2010). Thus one of the objectives of the Source Term (ST) work package has been to provide information on the behaviour of fission products (FPs), such as ruthenium, under highly oxidizing conditions. In case of a severe accident in a nuclear power plant (NPP), when the coolant is lost, fuel will rapidly heat-up leading to a release of some of the fission products that have been generated and are located in its micro-structure. If air gets into contact with ruthenium in the fuel, the formation of volatile ruthenium oxides is probable. The radiotoxicity of these oxides is near that of iodine in the short term and near that of caesium in the long term. To limit the possible source term, it is of interest to understand the chemical behaviour of ruthenium oxides when they are transported through the reactor coolant system (RCS) into the containment atmosphere.

The transport of Ru in primary circuit conditions has been studied in separate-effect tests with small-scale experimental facilities as a part of SARNET (Backman et al., 2005; Kärkelä et al., 2007; Vér et al., 2010, 2012). Ruthenium was oxidized at high temperature under dry or humid atmosphere close to atmospheric pressure with specific attention being paid to the transport of Ru through a thermal gradient tube (TGT). The effect of other fission products and control rod materials on the transport of Ru was also investigated. The observed behaviour of ruthenium in the tests was analysed with CFD/FLUENT and ASTEC/SOPHAEROS simulations (Davidovich, 2008; Kärkelä et al., 2008; Demenay, 2012).

* Corresponding author.
E-mail address: Teemu.Karkela@vtt.fi (T. Kärkelä).

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The large-scale integral tests of the Phébus FP program (Clément and Zeyen, 2013) studied the degradation of real fuel bundle and the release, transport and deposition of FPs, e.g., ruthenium, as well as structural materials and control rod materials in the model of primary circuit and containment building. The tests were conducted under steam-rich or steam-poor atmospheres and under low pressures (~0.2 MPa).

This paper summarizes the main results of these experimental and simulation studies on the transport of Ru in reactor coolant system conditions. The results are discussed considering the effects on source term evaluations and the future experimental as well as modelling needs are highlighted.

2. Ruthenium behaviour in separate-effect tests

2.1. Ruthenium transport tests at VTT

The aim in the VTT ruthenium transport tests (Backman et al., 2005; Kärkelä et al., 2007) was to study the transport and deposition of ruthenium in primary circuit conditions. The high temperature oxidation of Ru (827–1427 °C) in a mixture of air, steam and argon and the following Ru speciation and transport within a TGT, which outlet was at *ca.* 25 °C, were monitored both on-line and off-line.

2.1.1. Experimental facility and test conditions

The main component of the system for the tests (Fig. 1) was a horizontal tubular flow furnace, in which the ruthenium source (RuO_2 powder or gaseous RuO_4) was heated in an oxidizing flow (5 Nl/min, 0 °C, 101,325 Pa). The furnace (Entech, ETF20/18-II-L) used was 110 cm long. The tube material in the furnace was high purity alumina. The ceramic crucible with the RuO_2 powder (mass 1 g) was placed over the second heated zone of the furnace.

As the gas exited the furnace, it cooled in a stainless steel (SS; AISI 316L) tube and the gaseous ruthenium oxides decomposed partly to solid RuO_2. Aerosol particles were filtered out at point 106 cm downstream of the furnace. After the filter, gaseous ruthenium was trapped in a 1 M NaOH-water solution, cooled in an ice-bath. Aerosol gas-phase sampling was done at point 74 cm. The sample was diluted and quenched to *ca.* 25 °C with a porous tube diluter in order to minimise losses during dilution. The number size distribution of the particles was measured with a combination of differential mobility analyser (DMA, TSI 3081) and condensation nucleus counter (CNC, TSI 3022).

In some tests gaseous RuO_4 was fed into the facility instead of vaporizing RuO_2 powder. The method to produce gaseous RuO_4 is described in Kärkelä et al. (2007). A system to feed gaseous RuO_4 was added to the inlet of the facility (Fig. 1) and aerosol sampling line was not connected. The first 64 cm of the stainless steel tube was replaced with alumina tube of the same length and diameter in some experiments.

2.1.2. Results of the VTT ruthenium transport tests

A significant difference in the decomposition rate of gaseous ruthenium depending on the furnace temperature was found (Kärkelä et al., 2007). At 827 °C the Ru transport rate values were rather uncertain though due to a very low release rate. At 1027 °C Ru was primarily transported in gaseous form. When air flow was saturated with water vapour at 30 °C the fraction of gaseous Ru of all ruthenium transported through the facility was as high as 89%. In dry air the gaseous Ru fraction decreased to 62%. At the same time, the partial pressure of RuO_4 at the circuit outlet was in the range of 10^{-6} to 10^{-7} bar. Only up to 6% of the released Ru was transported through the primary circuit model as particles.

At 1227 °C and above, the partial pressure of gaseous RuO_3 is clearly dominant according to thermodynamic equilibrium (Backman et al., 2005). This favours the reactive condensation and formation of RuO_2 particles inside the TGT. The transported aerosol fraction of all Ru released increased from 12% to 25% when temperature increased from 1227 to 1427 °C. Gaseous Ru seemed to react on the surface of the SS tube, on top of which RuO_2 particles had been deposited. Thus the fraction of solid ruthenium of all the Ru transported through the facility was higher than 99%. It became clear that Ru transport took place almost entirely as particles when Ru was oxidized at high temperatures. The partial pressure of the transported RuO_4 remained in the range of 10^{-6} to 10^{-8} bar. Furthermore, the effect of silver seed particles on the transport of Ru was investigated in a test at 1227 °C. The seed particles were produced from a precursor solution of silver nitrate ($AgNO_3$) and water. As the generated droplets of precursor solution were heated at the inlet of the furnace, silver nitrate decomposed to silver particles, oxygen (g) and nitrogen dioxide (g). At the same time water was vaporized. As a result, the transport of Ru in gaseous form through the TGT was 50 times higher than in tests under

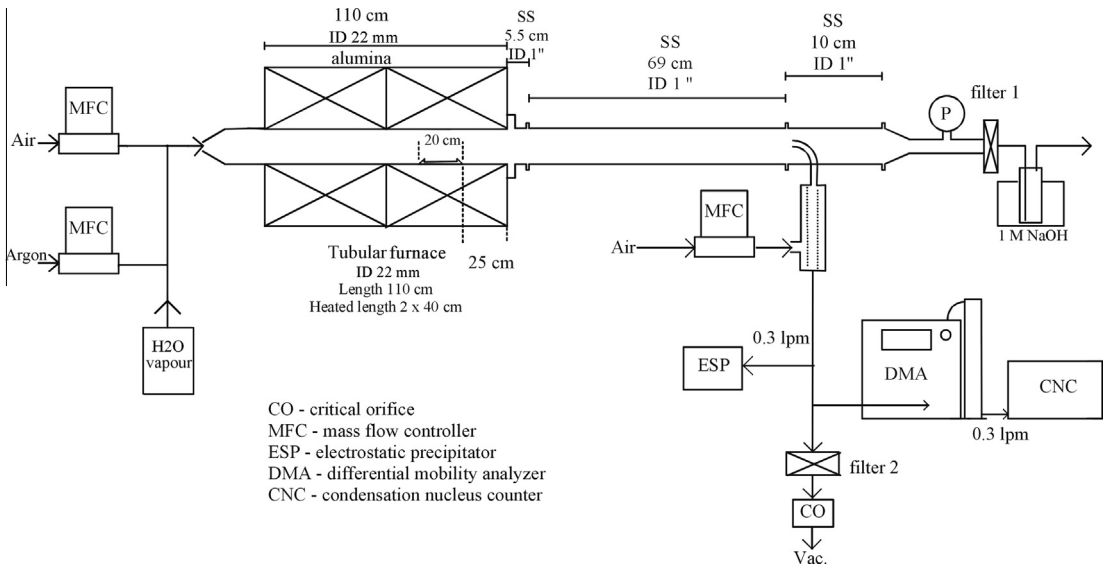

Fig. 1. Experimental set-up for ruthenium tests.