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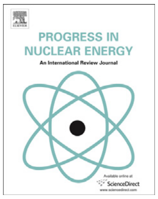

Ruthenium transport in an RCS with airborne CsI

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ABSTRACT

Ruthenium is one of the most radiotoxic fission products which can be released from fuel as ruthenium oxides in an air ingress accident at a nuclear power plant. In this study it was found that the transport of the released ruthenium oxides through a reactor coolant system into the containment building is significantly affected by the atmospheric conditions. Airborne CsI increased the transport of gaseous ruthenium compared with that in a pure air atmosphere. The overall transport of ruthenium increased with temperature. In order to understand the behaviour of ruthenium in accident conditions, it is important to widen the experimental conditions from pure air/steam atmospheres to more realistic mixtures of prototypic gases and aerosols.

1. Introduction

During normal operation of a nuclear power plant (NPP), fission products are produced in the fuel pellets, e.g. UO_2, as by-products of a neutron radiation-stimulated fission reaction. The produced fission products are retained within the fuel matrix and in a gas space between the cladding material and fuel pellet. In a severe nuclear power plant accident, when coolant is lost from the reactor and the reactor temperature increases, degradation of fuel pellets takes place eventually. As the integrity of fuel pellet cladding material is lost, release of fission products from the pellets is initiated and if the source term mitigation measures are insufficient, radiotoxic compounds may be transported into the environment. This causes a threat to the population, particularly to the employees of the NPP, as radiation may cause diseases, such as cancer, when citizens are exposed to fission products via deposition on the skin or when airborne fission products are inhaled and ingested or when the radiation dose rate in the environment is significantly increased.

Release of the fission product ruthenium from 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 (Haste et al., 2013; Grégoire and Haste, 2013) it was observed that most of the released ruthenium was transported to the containment building as solid RuO_2. Small-scale experiments (Backman et al., 2005; Kärkelä et al., 2007; Vér et al., 2012) have shown that the transport of gaseous ruthenium through a reactor coolant system (RCS) 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 high temperature gradient area of a model primary circuit (Kärkelä et al., 2014). As a result, the observed partial pressure of RuO_4 in containment conditions was at a level of 10^{-6} to 10^{-8} bar at 310 K–400 K. The research on air ingress conditions was taken even further in a recent study (Kajan et al., 2017a), 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.

In addition to gaseous additives, the gas flow in a reactor coolant

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Abbreviations

AS	aspiration sampler
CMD	count median diameter
CO	critical orifice
CPC	condensation particle counter
DMA	differential mobility analyzer
ELPI	electrical low pressure impactor
HPGe	high purity germanium detector
ICP-MS	inductively coupled plasma mass spectrometry
INAA	instrumental neutron activation analysis
MFC	mass flow controller
NPP	nuclear power plant
PTFE	polytetrafluoroethylene
RCS	reactor coolant system
SEM	scanning electron microscope
SMPS	scanning mobility particle sizer
T/P	temperature and pressure
XPS	X-ray photoelectron spectroscopy
ρ_0	reference density (1 g/cm3)
ρ_p	particle density
C_s (d_a)	Cunningham slip correction factor for the aerodynamic diameter
C_s (d_m)	Cunningham slip correction factor for the mobility diameter
C_s (d_ve)	Cunningham slip correction factor for the volume equivalent diameter
d_a	aerodynamic diameter
d_m	(electrical) mobility diameter
d_ve	volume equivalent diameter
χ	dynamic shape factor

system in severe accident conditions also includes aerosols, formed by 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 (Kajan et al., 2017b), 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 (Clément et al., 2007). There is some evidence of the retention of ruthenium on a surface coated with caesium at high temperature (750–900 K) (Vér et al., 2010) and of the trapping of gaseous ruthenium by CsI deposit at low temperature (ca. 300 K) (Kärkelä et al., 2007). 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.

2. Experimental

2.1. Experimental facility

The configuration of the “VTT Ru transport facility” used in these experiments is presented in Fig. 1. A detailed description of the facility was presented in the previous studies (Kärkelä et al., 2007; Kajan et al., 2017a). The main component of the facility was the horizontal, tubular flow furnace (Entech, ETF20/18-II-L), which was used to heat the anhydrous RuO_2 powder (99.95%, Alfa Aesar). The furnace was 110 cm long and it had two heating sections, each 40 cm long. These zones were separated by a 38 mm layer of insulation. At both ends of the furnace there was a 131 mm thermal insulation. The furnace tube was made of high purity alumina (Al_2O_3, 99.7%) and its inner diameter was 22 mm. The alumina crucible with the RuO_2 powder (mass ca. 1 g) was placed in the middle of the second heated zone of the furnace. The RuO_2 powder was heated to 1300 K, 1500 K or 1700 K in an oxidizing flow in order to produce gaseous ruthenium oxides.

The total air flow rate through the facility was 5.0 ± 0.1 l/min (NTP; NTP conditions 0 °C, 101325 Pa, measured with a Thermal Mass Flowmeter TSI 3063, TSI Incorp.). The pressure inside the facility ranged from 102 to 104 kPa. The air flow directed to the furnace tube was either dry or humid. In the case of dry atmosphere, the air flow of 5.0 ± 0.1 l/min (NTP) was directed to the furnace. In the case of humid atmosphere, the air flow 2.50 ± 0.05 l/min (NTP) was fed through an atomizer (TSI 3076) and the flow transported the droplets (Milli-Q, ultrapure water, resistivity 18.2 MΩ cm at 25 °C) produced by the atomizer via the heated line (120 °C) to the furnace. The gas flow was mixed with another air flow 2.50 ± 0.05 l/min (NTP) before the inlet of furnace. Water evaporated when the droplets were heated, thus it led to an increase in the steam concentration within the furnace. CsI (99.999%, Sigma-Aldrich) was fed to the facility by mixing CsI powder with ultrapure water in the supply bottle of the atomizer and the generated droplets transported CsI to the furnace tube. As water evaporated from the droplets, the formation of solid CsI particles occurred.

After the vaporization of Ru and the subsequent reactions within the gaseous atmosphere, the gaseous and particulate reaction products were trapped in NaOH solution and collected on plane polytetrafluoroethylene (PTFE) filter (see “filter 1” in Fig. 1), respectively, at the outlet of the facility where temperature had decreased to ca. 300 K. Particle samples were also collected on perforated carbon-coated nickel (400 mesh) grids with the aspiration sampler. An additional PTFE filter (see “filter 2” in Fig. 1) was placed into the aerosol online analysis line to enable particle chemical analysis after the experiment, see below. A detailed description of the facility is given in (Kajan et al., 2017b). The details of particle online analysis are given below.

2.2. Experimental procedure and matrix

The experiments were started with placing a crucible filled with RuO_2 powder (1 g) into the furnace and then heating up the system (heating rate of 10 °C per minute). A nitrogen gas flow of 0.50 ± 0.01 l/min (NTP) was directed through the facility during the heating up phase in order to flush out the oxygen in the system. The main gas flow through the facility was started when the furnace set-point was reached. In the experiments, particulate and gaseous reaction products were collected on filter and trapped in a 1 M NaOH solution, respectively. At the same time, particles in the gas phase were analysed online and additional samples of the particles were collected for the analyses to be conducted later. After the experiment, the gas flows were stopped and the filters and trapping solutions were removed. The facility was sealed airtight. When the facility was cooled down after several hours (cooling rate of 10 °C per minute), the crucible was removed.

The experimental matrix is presented in Table 1. Experiments 1 to 3 were conducted in pure dry or humid air atmosphere without aerosol additives. These experiments are also considered as reference experiments for experiments 4 to 6, which were conducted in